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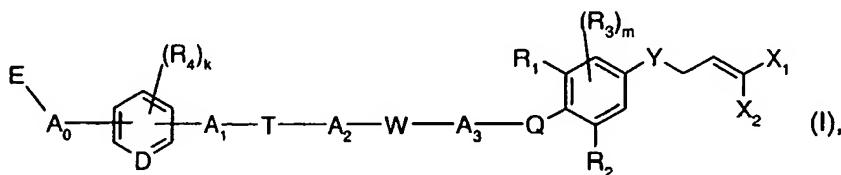
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(54) Title: 4-(3,3-DIHALO-ALLYLOXY) PHENOL DERIVATIVES HAVING PESTICIDAL PROPERTIES

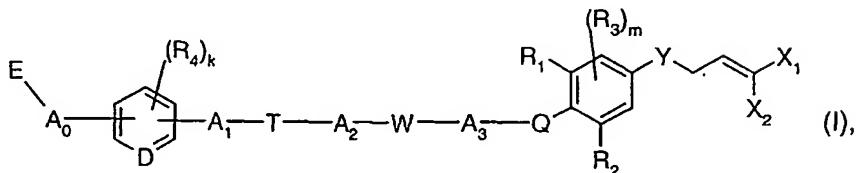


(57) Abstract: Compounds of formula (I) wherein A₀ is a bond or a C₁-C₆alkylene bridge; A₁, A₂ and A₃ are a C₁-C₆alkylene bridge; D is CH or N; W is, for example, O, S, SO, SO₂, -C(=O)-O- or -O-C(=O)-; T is a bond or, for example, O, NH, S, SO or SR₂; Q is, for example, O, S, SO or SO₂; Y is, for example, O, S, SO or SO₂; X₁ and X₂ are each independently of the other fluorine, chlorine or bromine; R₁, R₂ and R₃ are, for example, H, halogen, OH, SH, CN or nitro; R₄ is, for example, H, halogen, OH, SH, CN, nitro or C₁-C₆alkyl; R₅ is, for example, H, CN, OH, C₁-C₆alkyl or C₃-C₈cycloalkyl; k is 1, 2 or 3 when D is nitrogen; or is 1, 2, 3 or 4 when D is CH; m is 1 or 2; E is heteroaryl which is unsubstituted or substituted - depending upon the substitutions possible on the ring - by from one to four identical or different substituents selected from R₁₀; and R₁₀ is, for example, halogen, CN, NO₂, OH, SH or C₁-C₆alkyl; and, where applicable, their possible E/Z isomers, E/Z isomeric mixtures and/or tautomers, in each case in free form or in salt form, a process for the preparation of those compounds and their use, pesticidal compositions in which the active ingredient has been selected from those compounds or an agrochemically acceptable salt thereof, a process for the preparation of those compositions and their use, plant propagation material treated with those compositions, and a method of controlling pests.

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4-(3,3-Dihalo-allyloxy) Phenol Derivatives having pesticidal properties

The present invention relates (1) to compounds of formula



wherein

A₀, A₁ and A₂ are each independently of the other a bond or a C₁-C₆alkylene bridge which is unsubstituted or substituted by from one to six identical or different substituents selected from C₃-C₈cycloalkyl, C₃-C₈cycloalkyl-C₁-C₆alkyl and C₁-C₃haloalkyl;

A₃ is a C₁-C₆alkylene bridge which is unsubstituted or substituted by from one to six identical or different substituents selected from C₃-C₈cycloalkyl, C₃-C₈cycloalkyl-C₁-C₆alkyl and C₁-C₃haloalkyl;

D is CH or N;

X₁ and X₂ are each independently of the other fluorine, chlorine or bromine;

R₁, R₂ and R₃ are each independently of the others H, halogen, OH, SH, CN, nitro, C₁-C₆alkyl, C₁-C₆haloalkyl, C₁-C₆alkylcarbonyl, C₂-C₆alkenyl, C₂-C₆haloalkenyl, C₂-C₆alkynyl, C₁-C₆alkoxy, C₁-C₆haloalkoxy, C₂-C₆alkenyloxy, C₂-C₆haloalkenyloxy, C₂-C₆alkynyoxy, -S(=O)-C₁-C₆alkyl, -S(O)₂-C₁-C₆alkyl, C₁-C₆alkoxycarbonyl or C₃-C₆haloalkynyoxy; the substituents R₃ being independent of one another when m is 2;

R₄ is H, halogen, OH, SH, CN, nitro, C₁-C₆alkyl, C₁-C₆haloalkyl, C₁-C₆alkylcarbonyl, C₂-C₆alkenyl, C₂-C₆haloalkenyl, C₂-C₆alkynyl, C₁-C₆alkoxy, C₁-C₆haloalkoxy, C₂-C₆alkenyl-oxy, C₂-C₆haloalkenyl-oxy, C₂-C₆alkynyoxy, -S(=O)-C₁-C₆alkyl, -S(=O)₂-C₁-C₆alkyl or C₁-C₆alkoxycarbonyl; the substituents R₄ being independent of one another when k is greater than 1; or N(R₅)₂ wherein the two substituents R₅ are independent of one another;

R₅ is H, CN, OH, C₁-C₆alkyl, C₃-C₈cycloalkyl, C₃-C₈cycloalkyl-C₁-C₆alkyl, C₁-C₆haloalkyl, C₁-C₆alkoxy, C₁-C₆haloalkoxy, C₂-C₆alkenyloxy, C₂-C₆haloalkenyloxy, C₂-C₆alkynyoxy, -C(=O)R₆, -C(=S)R₆, phenyl, benzyl; or phenyl or benzyl each of which is substituted in the aromatic ring by from one to five identical or different substituents selected from the group

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consisting of halogen, C₁-C₆alkyl, halo-C₁-C₆alkyl, C₁-C₆alkoxy, halo-C₁-C₆alkoxy, hydroxy, cyano and nitro;

or the two substituents R₅ together form a four- to eight-membered, straight-chain or branched alkylene bridge wherein a CH₂ group may have been replaced by O, S or NR₉, and the alkylene bridge is unsubstituted or substituted by from one to four identical or different substituents selected from C₃-C₈cycloalkyl, C₃-C₈cycloalkyl-C₁-C₆alkyl and C₁-C₃haloalkyl;

W is O, NR₆, S, SO, SO₂, -C(=O)-O-, -O-C(=O)-, -C(=O)-NR₇- or -NR₇-C(=O)-;

T is a bond, O, NH, NR₆, S, SO, SO₂, -C(=O)-O-, -O-C(=O)-, -C(=O)-NR₇- or -NR₇-C(=O)-;

Q is O, NR₆, S, SO or SO₂;

Y is O, NR₆, S, SO or SO₂;

R₆ and R₇ are independently of each other H, C₁-C₆alkyl, C₁-C₃haloalkyl, C₁-C₆alkyl-carbonyl, C₁-C₃haloalkylcarbonyl, C₁-C₆alkoxyalkyl, C₃-C₈cycloalkyl or benzyl;

R₈ is C₁-C₆alkyl, C₁-C₆haloalkyl, C₂-C₆alkenyl, C₂-C₆haloalkenyl, C₂-C₆alkynyl, C₁-C₆alkoxy, C₁-C₆haloalkoxy, C₂-C₆alkenyloxy, C₂-C₆haloalkenyloxy, C₂-C₆alkynyloxy, C₃-C₈cycloalkyl, phenyl, benzyl; or phenyl or benzyl each of which is unsubstituted or substituted by from one to three identical or different substituents selected from halogen, CN, nitro, C₁-C₆alkyl, C₁-C₆haloalkyl, C₁-C₆alkylcarbonyl, C₂-C₆alkenyl, C₂-C₆haloalkenyl, C₂-C₆alkynyl, C₁-C₆alkoxy, C₁-C₆haloalkoxy, C₁-C₆alkoxycarbonyl, C₁-C₃haloalkoxycarbonyl and C₂-C₆haloalkenyloxy;

R₉ is H, C₁-C₆alkyl, C₁-C₃haloalkyl, C₁-C₆-alkylcarbonyl, C₁-C₆haloalkylcarbonyl, C₁-C₆alkoxyalkyl, C₃-C₈cycloalkyl or benzyl;

k is 1, 2 or 3 when D is nitrogen; or is 1, 2, 3 or 4 when D is CH;

m is 1 or 2;

E is heteroaryl which is unsubstituted or substituted - depending upon the substitutions possible on the ring - by from one to four identical or different substituents selected from R₁₀;

R₁₀ is halogen, CN, NO₂, OH, SH, C₁-C₆alkyl, C₁-C₆haloalkyl, C₁-C₆hydroxyalkyl, C₃-C₈cycloalkyl, C₃-C₈cycloalkyl-C₁-C₆alkyl, C₂-C₆alkenyl, C₂-C₆haloalkenyl, C₃-C₆alkynyl, C₃-C₆haloalkynyl, C₁-C₆alkoxy, C₁-C₆alkoxy-C₁-C₆alkyl, C₁-C₆haloalkoxy, C₁-C₆haloalkoxy-C₁-C₆alkyl, C₂-C₆alkenyloxy, C₂-C₆haloalkenyloxy, C₂-C₆alkenyloxy-C₁-C₆alkyl, C₂-C₆haloalkenyloxy-C₁-C₆alkyl, C₃-C₆alkynyl, C₃-C₆haloalkynyl, C₃-C₆alkoxyalkyl, C₃-C₆haloalkoxyalkyl, C₃-C₆alkoxyalkoxy, C₃-C₆haloalkoxyalkoxy, C₃-C₆alkoxyalkynyl, C₃-C₆haloalkoxyalkynyl, C₃-C₆alkynylalkyl, C₃-C₆haloalkynylalkyl, C₃-C₆alkynylalkoxy, C₃-C₆haloalkynylalkoxy, C₃-C₆alkynylalkoxyalkyl, C₃-C₆haloalkynylalkoxyalkyl.

$C_3\text{-}C_8\text{cycloalkoxy}$, $C_3\text{-}C_8\text{cycloalkyl-C}_1\text{-}C_6\text{alkoxy}$, $C_3\text{-}C_8\text{cycloalkoxy-C}_1\text{-}C_6\text{alkyl}$, $C_3\text{-}C_8\text{cyclo-alkoxy-C}_1\text{-}C_6\text{alkoxy}$, $C_3\text{-}C_8\text{cycloalkyl-C}_1\text{-}C_6\text{alkoxy-C}_1\text{-}C_6\text{alkyl}$, $C_1\text{-}C_6\text{alkylcarbonyl-C}_1\text{-}C_6\text{alkyl}$, $C_1\text{-}C_6\text{alkoxycarbonyl-C}_1\text{-}C_6\text{alkyl}$, $C_1\text{-}C_6\text{alkylthio}$, $C_2\text{-}C_6\text{alkenylthio}$, $C_3\text{-}C_6\text{alkynylthio}$, $C_3\text{-}C_6\text{cycloalkylthio}$, $C_3\text{-}C_6\text{cycloalkyl-C}_1\text{-}C_6\text{alkylthio}$, $C_2\text{-}C_6\text{haloalkenylthio}$, $C_1\text{-}C_6\text{haloalkylthio}$, NH_2 , $NH(C_1\text{-}C_6\text{alkyl})_2$, $C_1\text{-}C_6\text{alkylcarbonylamino}$, $C_1\text{-}C_6\text{haloalkylcarbonylamino}$, $C_1\text{-}C_6\text{alkoxycarbonylamino}$, $C_1\text{-}C_6\text{alkylaminocarbonylamino}$, $\text{-SO-C}_1\text{-}C_6\text{alkyl}$, $\text{-SO-halo-C}_1\text{-}C_6\text{alkyl}$, $\text{-SO}_2\text{-C}_1\text{-}C_6\text{alkyl}$, $\text{-SO}_2\text{-halo-C}_1\text{-}C_6\text{alkyl}$, -C(=O)R_{11} , phenyl or benzyl; wherein the phenyl and benzyl radicals may be unsubstituted or may carry independently of each other one to three substituents selected from the group consisting of halogen, OH, SH, CN, nitro, $C_1\text{-}C_6\text{alkyl}$, $C_1\text{-}C_6\text{haloalkyl}$, $C_1\text{-}C_6\text{alkylcarbonyl}$, $C_2\text{-}C_6\text{alkenyl}$, $C_2\text{-}C_6\text{haloalkenyl}$, $C_2\text{-}C_6\text{alkynyl}$, $C_1\text{-}C_6\text{alkoxy}$, $C_1\text{-}C_6\text{haloalkoxy}$, $C_2\text{-}C_6\text{alkenyloxy}$, $C_2\text{-}C_6\text{haloalkenyloxy}$, $C_2\text{-}C_6\text{alkynyoxy}$, $\text{-S(=O)-C}_1\text{-}C_6\text{alkyl}$, $\text{-S(O)}_2\text{-C}_1\text{-}C_6\text{alkyl}$, $C_1\text{-}C_6\text{alkoxycarbonyl}$ and $C_2\text{-}C_6\text{haloalkenyloxy}$; and

R_{11} is H, OH, $C_1\text{-}C_6\text{alkyl}$, $C_3\text{-}C_8\text{cycloalkyl}$, $C_3\text{-}C_8\text{cycloalkyl-C}_1\text{-}C_6\text{alkyl}$, $C_1\text{-}C_6\text{haloalkyl}$, $C_1\text{-}C_6\text{alkoxy}$, $C_3\text{-}C_8\text{cycloalkoxy}$, $C_3\text{-}C_8\text{cycloalkyl-C}_1\text{-}C_6\text{alkoxy}$, $C_1\text{-}C_6\text{haloalkoxy}$, $C_2\text{-}C_6\text{alkenyl}$, $C_2\text{-}C_6\text{haloalkenyl}$, $C_2\text{-}C_6\text{alkenyloxy}$, $C_2\text{-}C_6\text{haloalkenyloxy}$, $C_2\text{-}C_6\text{alkynyl}$, $C_2\text{-}C_6\text{haloalkynyl}$, $C_2\text{-}C_6\text{alkynyoxy}$, $C_2\text{-}C_6\text{haloalkynyoxy}$, NH_2 , $NH(C_1\text{-}C_6\text{alkyl})_2$, NH-phenyl, NH-benzyl, phenoxy or benzyloxy;

and, where applicable, their possible E/Z isomers, E/Z isomeric mixtures and/or tautomers, in each case in free form or in salt form, to a process for the preparation of those compounds, E/Z isomers and tautomers and to their use, to pesticidal compositions in which the active ingredient has been selected from those compounds, E/Z isomers and tautomers, and to a process for the preparation of those compositions and to their use, to intermediates and, where applicable, their possible E/Z isomers, E/Z isomeric mixtures and/or tautomers, in free form or in salt form, for the preparation of those compounds, where applicable to tautomers, in free form or in salt form, of those intermediates and to a process for the preparation of those intermediates and their tautomers and to their use.

Certain dihaloallyl derivatives are proposed in the literature as active ingredients in pesticidal compositions. The biological properties of those known compounds are not entirely satisfactory in the field of pest control, however, for which reason there is a need to provide further compounds having pesticidal properties, especially for controlling insects and members of the order Acarina, that problem being solved according to the invention by the provision of the present compounds of formula (I).

The compounds of formula (I) and, where applicable, their tautomers are able to form salts, e.g. acid addition salts. The latter are formed, for example, with strong inorganic acids, such as mineral acids, e.g. sulfuric acid, a phosphoric acid or a hydrohalic acid, with strong organic carboxylic acids, such as unsubstituted or substituted, e.g. halo-substituted, C₁-C₄ alkanecarboxylic acids, for example acetic acid, saturated or unsaturated dicarboxylic acids, e.g. oxalic, malonic, maleic, fumaric or phthalic acid, hydroxycarboxylic acids, e.g. ascorbic, lactic, malic, tartaric or citric acid, or benzoic acid, or with organic sulfonic acids, such as unsubstituted or substituted, e.g. halo-substituted, C₁-C₄ alkane- or aryl-sulfonic acids, e.g. methane- or p-toluene-sulfonic acid. Furthermore, compounds of formula (I) having at least one acid group are able to form salts with bases. Suitable salts with bases are, for example, metal salts, such as alkali metal or alkaline earth metal salts, e.g. sodium, potassium or magnesium salts, or salts with ammonia or an organic amine, such as morpholine, piperidine, pyrrolidine, a mono-, di- or tri-lower alkylamine, e.g. ethyl-, diethyl-, triethyl- or dimethyl-propyl-amine, or a mono-, di- or tri-hydroxy-lower alkylamine, e.g. mono-, di- or tri-ethanolamine. It may also be possible for corresponding internal salts to be formed. The free form is preferred. Of the salts of compounds of formula (I), preference is given to agro-chemically advantageous salts. Hereinabove and hereinbelow any reference to the free compounds of formula (I) or to their salts is to be understood as including, where appropriate, also the corresponding salts or the free compounds of formula (I), respectively. The same applies to tautomers of compounds of formula (I) and their salts.

The general terms used hereinabove and hereinbelow have the meanings given below, unless defined otherwise.

Halogen, as a group *per se* and as a structural element of other groups and compounds, such as haloalkyl, halocycloalkyl, haloalkenyl, haloalkynyl and haloalkoxy, is fluorine, chlorine, bromine or iodine, especially fluorine, chlorine or bromine, more especially fluorine or chlorine, especially chlorine.

Unless defined otherwise, carbon-containing groups and compounds each contain from 1 up to and including 20, preferably from 1 up to and including 18, especially from 1 up to and including 10, more especially from 1 up to and including 6, especially from 1 up to and including 4, especially from 1 up to and including 3, more especially 1 or 2, carbon atoms; methyl is especially preferred.

Alkylene is a straight-chain or branched bridging member and is especially -CH₂-, -CH₂CH₂-, -CH₂-CH₂-CH₂-, -CH₂-CH₂-CH₂-CH₂-, -CH₂-CH₂-CH₂-CH₂-CH₂-, -CH(CH₃)-,

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-CH₂(CH₃)CH₂-CH₂-, -CH(C₂H₅)-, -C(CH₃)₂-, -CH(CH₃)CH₂-, -CH(CH₃)CH(CH₃)- or -CH₂C(CH₃)₂-CH₂-.

Alkyl, as a group *per se* and as a structural element of other groups and compounds, such as haloalkyl, alkoxy, alkoxyalkyl, haloalkoxy, alkoxy carbonyl, alkylthio, haloalkylthio, alkylsulfonyl and alkylsulfonyloxy, is - in each case giving due consideration to the number of carbon atoms contained in the group or compound in question - either straight-chain, e.g. methyl, ethyl, n-propyl, n-butyl, n-hexyl, n-octyl, n-decyl, n-dodecyl, n-hexadecyl or n-octadecyl, or branched, e.g. isopropyl, isobutyl, sec-butyl, tert-butyl, isopentyl, neopentyl or isohexyl.

Alkenyl and alkynyl - as groups *per se* and as structural elements of other groups and compounds, such as haloalkenyl, haloalkynyl, alkenyloxy, haloalkenyloxy, alkynyoxy or haloalkynyoxy - are straight-chain or branched and each contains two or preferably one unsaturated carbon-carbon bond(s). There may be mentioned by way of example vinyl, prop-2-en-1-yl, 2-methylprop-2-en-1-yl, but-2-en-1-yl, but-3-en-1-yl, prop-2-yn-1-yl, but-2-yn-1-yl and but-3-yn-1-yl.

Cycloalkyl - as a group *per se* and as a structural element of other groups and compounds, such as alkyl - is cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl, cycloheptyl or cyclooctyl. Cyclopentyl and cyclohexyl, and especially cyclopropyl, are preferred.

Halo-substituted carbon-containing groups and compounds, such as haloalkyl and haloalkoxy, may be partially halogenated or perhalogenated, the halogen substituents in the case of polyhalogenation being the same or different. Examples of haloalkyl - as a group *per se* and as a structural element of other groups and compounds, such as haloalkoxy - are methyl substituted from one to three times by fluorine, chlorine and/or bromine, such as CHF₂, CF₃ or CH₂Cl; ethyl substituted from one to five times by fluorine, chlorine and/or bromine, such as CH₂CF₃, CF₂CF₃, CF₂CCl₃, CF₂CHCl₂, CF₂CHF₂, CF₂CFCl₂, CH₂CH₂Cl, CF₂CHBr₂, CF₂CHClF, CF₂CHBrF or CCIFCHClF; propyl or isopropyl each substituted from one to seven times by fluorine, chlorine and/or bromine, such as CH₂CHBrCH₂Br, CF₂CHFCF₃, CH₂CF₂CF₃, CF₂CF₂CF₃, CH(CF₃)₂ or CH₂CH₂CH₂Cl; and butyl or an isomer thereof substituted from one to nine times by fluorine, chlorine and/or bromine, such as CF(CF₃)CHFCF₃, CF₂(CF₂)₂CF₃ or CH₂(CF₂)₂CF₃.

Aryl is especially phenyl or naphthyl, preferably phenyl.

Heteroaryl is to be understood as meaning a five- to seven-membered, especially a five-membered, monocyclic aromatic ring which contains from one to four hetero atoms selected from the group consisting of N, O and S, especially N and S, or a bicyclic heteroaryl which may contain, either in one ring only - as in quinolinyl, quinoxaliny, indolinyl, benzothiophenyl or benzofuranyl, for example - or in both rings - as in pteridinyl or purinyl, for example - one or more hetero atoms selected independently of one another from N, O and S. Preference is given to pyridyl, pyrimidyl, pyrazinyl, pyridazinyl, s-triazinyl, 1,2,4-triazinyl, thienyl, furanyl, pyranyl, pyrrolyl, pyrazolyl, imidazolyl, thiazolyl, 1,2,3-triazolyl, 1,2,4-triazolyl, oxazolyl, isoxazolyl, thiadiazolyl, oxadiazolyl, tetrazolyl, benzothienyl, quinolinyl, quinoxaliny, benzofuranyl, benzimidazolyl, benzpyrrolyl, benzthiazolyl, indolyl, coumarinyl, or indazolyl, which are preferably bonded *via* a carbon atom;

special preference is given to thienyl, pyrrolyl, pyrazolyl, imidazolyl, 1,3-thiazolyl, 1,2,3-triazolyl, 1,2,4-triazolyl, 1,3,4-thiadiazolyl, 1,2,4-thiadiazolyl, oxazolyl, isoxazolyl, 1,2,4-oxadiazolyl or tetrazolyl.

Preferred embodiments within the scope of the invention are

- (2) compounds of formula (I) according to (1), wherein X₁ and X₂ are chlorine or bromine, especially chlorine;
- (3) compounds of formula (I) according to (1) or (2), wherein the group A₁-T-A₂ is a bond;
- (4) compounds of formula (I) according to (1) to (3), wherein W is oxygen, -C(=O)O- or -C(=O)NH-, especially O;
- (5) compounds of formula (I) according to (1) to (4), wherein A₃ is a straight-chain alkylene bridge, especially ethylene, propylene or butylene, more especially propylene;
- (6) compounds of formula (I) according to (1) to (5), wherein Q is oxygen;
- (7) compounds of formula (I) according to (1) to (6), wherein Y is oxygen;
- (8) compounds of formula (I) according to (1) to (7), wherein R₁ and R₂ are bromine or chlorine, especially chlorine;
- (9) compounds of formula (I) according to (1) to (8), wherein R₃ is hydrogen;
- (10) compounds of formula (I) according to (1) to (9), wherein R₄ is hydrogen;

(11) compounds of formula (I) according to (1) to (10), wherein R₅ is H, C₁-C₆alkyl, C₃-C₈cycloalkyl, C₃-C₈cycloalkyl-C₁-C₆alkyl, C₁-C₆haloalkyl, C₁-C₆alkoxy, C₁-C₆haloalkoxy, C₂-C₆alkenyloxy, C₂-C₆haloalkenyloxy, C₂-C₆alkynyloxy;

(12) compounds of formula (I) according to (1) to (11), wherein R₆ is -C(=O)R₈ or -C(=S)R₈, and R₈ is C₁-C₆alkyl, C₁-C₆haloalkyl, C₂-C₆alkenyl, C₂-C₆haloalkenyl, C₂-C₆alkynyl, C₁-C₆alkoxy, C₁-C₆haloalkoxy, C₂-C₆alkenyloxy, C₂-C₆haloalkenyloxy, C₂-C₆alkynyloxy, C₃-C₆cycloalkyl or phenyl;

(13) compounds of formula (I) according to (1) or (2) and (4) to (11), wherein A₁ is a bond, T is oxygen and A₂ is a C₁-C₆alkylene bridge;

(14) compounds of formula (I) according to (1) or (2), wherein A₁ is a bond, T is -C(=O)O- wherein the oxygen is bonded to A₂, or is -C(=O)NH- wherein NH is bonded to A₂, and A₂ is a C₁-C₆alkylene bridge;

(15) compounds of formula (I) according to (1) to (14), wherein E is a five- to seven-membered, monocyclic heteroaryl having from one to four hetero atoms selected from the group consisting of N, O and S, or bicyclic heteroaryl which may contain, either in one ring only or in both rings, one or more hetero atoms selected independently of one another from N, O and S, and which is unsubstituted or carries one or two substituents R₁₀, which are independent of each other; especially which is unsubstituted or carries one substituent R₁₀;

especially five-membered monocyclic heteroaryl having from two to four hetero atoms selected from the group consisting of N, O and S;

more especially five-membered monocyclic heteroaryl having one or two nitrogen atoms and one oxygen atom;

also more especially five-membered monocyclic heteroaryl having three or four nitrogen atoms;

(16) compounds of formula (I) according to (1) to (15), wherein R₁₀ is H, CN, OH, NO₂, SH, C₁-C₆alkyl, C₁-C₆haloalkyl, C₂-C₆alkenyl, C₁-C₆alkoxy, C₁-C₆alkylcarbonyl, C₁-C₆alkoxycarbonyl, C₁-C₆alkylthio, C₁-C₆haloalkoxy, C₂-C₆alkenyloxy, C₂-C₆alkynyloxy, C₂-C₆alkenylthio, C₃-C₈cycloalkyl, C₃-C₈cycloalkyl-C₁-C₆alkyl, cycloalkyl-C₁-C₆alkoxy or cycloalkyl-C₁-C₆alkylthio;

especially C₁-C₆alkyl, C₁-C₆haloalkyl, C₂-C₆alkenyl, C₁-C₆alkoxy, C₁-C₆alkylthio, C₂-C₆-alkenyloxy, C₂-C₆alkenylthio, C₃-C₆cycloalkyl, cycloalkyl-C₁-C₆alkoxy or cycloalkyl-C₁-C₆-alkylthio;

(17) compounds of formula (I) according to (1) to (15), wherein E is tetrazol-1-yl which is unsubstituted or monosubstituted by C₁-C₆alkyl, C₁-C₆haloalkyl, C₂-C₆alkenyl, C₂-C₆haloalkenyl, C₃-C₆cycloalkyl-C₁-C₆alkyl, C₁-C₆alkoxy, C₁-C₆haloalkoxy, C₂-C₆alkenyloxy, C₂-C₆haloalkenyloxy, C₃-C₆alkynyloxy, C₃-C₆cycloalkyl-C₁-C₆alkoxy, C₁-C₆alkylthio, C₁-C₆haloalkylthio, C₂-C₆alkenylthio, C₂-C₆haloalkenylthio, C₃-C₆alkynylthio or by C₃-C₆cycloalkyl-C₁-C₆alkylthio;

(18) compounds of formula (I) according to (1) to (15), wherein E is tetrazol-5-yl which is unsubstituted or monosubstituted in the 2-position by C₁-C₆alkyl, C₁-C₆haloalkyl, C₂-C₆alkenyl, C₂-C₆haloalkenyl or by C₃-C₆cycloalkyl-C₁-C₆alkyl ;

(19) compounds of formula (I) according to (1) to (15), wherein E is 1,2,4-oxadiazol-3-yl which is unsubstituted or monosubstituted by CN, halogen, C₁-C₆alkyl, C₁-C₆haloalkyl, C₂-C₆alkenyl, C₂-C₆haloalkenyl, C₃-C₆alkynyl, C₃-C₆cycloalkyl, C₁-C₆alkylcarbonyl, C₁-C₆haloalkylcarbonyl or by C₁-C₆alkoxycarbonyl;

(20) compounds of formula (I) according to (1) to (15), wherein E is isoxazol-3-yl which is unsubstituted or monosubstituted by halogen, C₁-C₆alkyl, C₁-C₆haloalkyl, C₁-C₆alkoxy-methyl, C₁-C₆haloalkoxy-methyl, C₁-C₆haloalkylcarbonyloxy-methyl, C₁-C₆alkylcarbonyl, C₁-C₆haloalkylcarbonyl or by C₁-C₆alkoxycarbonyl;

(21) compounds of formula (I) according to (1) to (15), wherein E is thiazol-2-yl which is unsubstituted or mono- or disubstituted by halogen, OH, CN, C₁-C₆alkyl, C₁-C₆haloalkyl, C₃-C₆cycloalkyl, C₁-C₆alkylcarbonyl, C₁-C₆haloalkylcarbonyl or by C₁-C₆alkoxycarbonyl, and wherein the substituents are independent of each other;

(22) compounds of formula (I) according to (1) to (15), wherein E is 1,3,4-thiadiazol-2-yl which is unsubstituted or monosubstituted by halogen, CN, C₁-C₆alkyl, C₁-C₆haloalkyl, C₂-C₆alkenyl, C₂-C₆haloalkenyl, C₃-C₆cycloalkyl-C₁-C₆alkyl, C₁-C₆alkoxy, C₁-C₆haloalkoxy, C₂-C₆alkenyloxy, C₂-C₆haloalkenyloxy, C₃-C₆alkynyloxy, C₃-C₆cycloalkyl-C₁-C₆alkoxy, C₁-C₆alkylthio, C₁-C₆haloalkylthio, C₂-C₆alkenylthio, C₂-C₆haloalkenylthio, C₃-C₆alkynylthio or by C₃-C₆cycloalkyl-C₁-C₆alkylthio;

(23) compounds of formula (I) according to (1) to (15), wherein E is 1,2,4-triazol-3-yl which is unsubstituted or substituted on the nitrogen in the 1-, 2- or 4-position by C₁-C₆alkyl

and is unsubstituted or substituted in the 5-position by halogen, NO₂, C₁-C₆alkyl, C₁-C₆-haloalkyl, C₂-C₆alkenyl, C₂-C₆haloalkenyl, C₃-C₆cycloalkyl-C₁-C₆alkyl, C₁-C₆alkoxy, C₁-C₆-haloalkoxy, C₂-C₆alkenyloxy, C₂-C₆haloalkenyloxy, C₃-C₆alkynyoxy, C₃-C₆cycloalkyl-C₁-C₆-alkoxy, C₁-C₆alkylthio, C₁-C₆haloalkylthio, C₂-C₆alkenylthio, C₂-C₆haloalkenylthio, C₃-C₆alkynylthio, C₃-C₆cycloalkyl-C₁-C₆alkylthio, NH₂, NHC(=O)C₁-C₆alkyl, NHC(=O)C₁-C₆haloalkyl or by NHC(=O)O-C₁-C₆alkyl;

(24) compounds of formula (I) according to (1) to (15), wherein E is thiazol-4-yl, which is mono- or disubstituted by halogen, OH, CN, C₁-C₆alkyl, C₁-C₆haloalkyl, C₃-C₆cycloalkyl, C₁-C₆alkylcarbonyl, C₁-C₆haloalkylcarbonyl or by C₁-C₆alkoxycarbonyl, and wherein the substituents are independent of each other;

(25) compounds of formula (I) according to (1) to (24), wherein D is CH;

(26) compounds of formula (I) according to (1) to (24), wherein D is N;

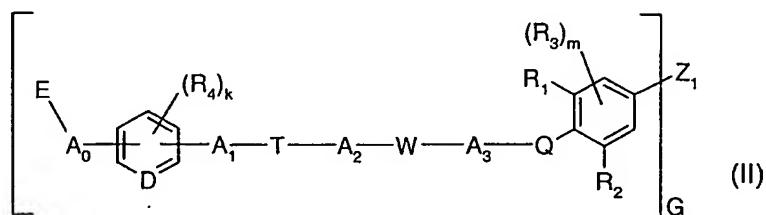
(27) compounds of formula (I) according to (1) to (26), wherein A₀ is -CH₂-;

(28) compounds of formula (I) according to (1) to (26), wherein A₀ is a bond.

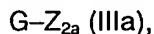
Special preference is given to the compounds listed in the Tables.

The invention relates also to a process for the preparation of a compound of formula (I), or a salt thereof, wherein

(a) a compound of formula



wherein A₀, A₁, A₂, A₃, D, E, W, Q, T, R₁, R₂, R₃, R₄, m and k are as defined for formula (I) under (1), Z₁ is -C(=O)R₁₂ and R₁₂ is H or C₁-C₆alkyl, is converted in the presence of an oxidising agent, especially a peracid, into a compound of formula



wherein Z_{2a} is O-C(=O)-R₁₃ and R₁₃ is C₁-C₆alkyl, and G denotes the part of the formula in brackets designated G in formula (II); either

(b) a compound of formula (IIIa) above or of formula

- 10 -

G-Z_{2b} (IIIb),

wherein G denotes the part of the formula in brackets designated G in formula (II), Z_{2b} is a radical of formula -Y-C(=O)R₁₄, Y is as defined for formula (I) under (1), and R₁₄ is C₁-C₁₂alkyl unsubstituted or substituted by from one to three identical or different halogen substituents, or is phenyl unsubstituted or substituted by from one to three identical or different substituents selected from halogen, CN, nitro, C₁-C₆alkyl, C₁-C₆haloalkyl, C₁-C₆alkylcarbonyl, C₂-C₆alkenyl, C₂-C₆haloalkenyl, C₂-C₆alkynyl, C₁-C₆alkoxy, C₁-C₆haloalkoxy, C₁-C₆alkoxycarbonyl and C₂-C₆haloalkenyloxy, is converted by hydrolytic cleavage into a compound of formula

G-Z₃ (IV),

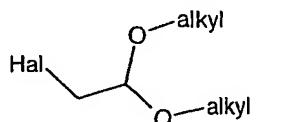
wherein G denotes the part of the formula in brackets designated G in formula (II), Z₃ is YH, and Y is as defined for formula (I) under (1); or

(c) a compound of formula

G-Z₄ (V),

wherein Z₄ is Y-CH₂-phenyl, wherein the phenyl radical is unsubstituted or substituted by from one to three identical or different substituents selected from halogen, CN, nitro, C₁-C₆alkyl, C₁-C₆haloalkyl, C₁-C₆alkylcarbonyl, C₂-C₆alkenyl, C₂-C₆haloalkenyl, C₂-C₆alkynyl, C₁-C₆alkoxy, C₁-C₆haloalkoxy, C₁-C₆alkoxycarbonyl and C₂-C₆haloalkenyloxy, G denotes the part of the formula in brackets designated G in formula (II), and Y is as defined for formula (I), is converted by removal of the benzyl group into a compound of formula (IV), as defined above;

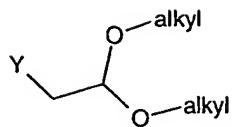
(d) the compound of formula (IV) so obtained is reacted in the presence of a base with a compound of formula



wherein Hal is halogen, preferably bromine or chlorine, and alkyl is C₁-C₆alkyl, or the two alkyl radicals together form a C₃-C₈alkylene bridge, to form a compound of formula

G-Z₅ (VI),

wherein G denotes the part of the formula in brackets designated G in formula (II), and Z₅ is



wherein alkyl and Y are as defined above;

(e) the compound of formula (VI) so obtained is converted by deprotection of the acetal function in the presence of an acid into a compound of formula

G-Z₆ (VII),

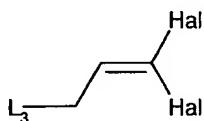
wherein Z₆ is a group $-Y-CH_2-C(=O)H$, G is as defined above for the compound of formula (II), and Y is as defined for formula (I) under (1), either

(f₁) for the preparation of a compound of formula (I) wherein X₁ and X₂ are chlorine or bromine, a compound of formula (VII) is reacted in the presence of a phosphine with a compound of formula C(X)₄ wherein X is chlorine or bromine; or

(f₂) for the preparation of a compound of formula (I) wherein X₁ and X₂ are chlorine, a compound of formula (VII) is reacted first with CCl₃-COOH or with chloroform in the presence of a strong base, then with acetic anhydride and subsequently with powdered zinc in acetic acid; or

(f₃) for the preparation of a compound of formula (I) wherein X₁ is fluorine and X₂ is chlorine or bromine, a compound of formula (VII) is reacted in the presence of a phosphine with a compound of the formula CF₂X₂, of the formula CFX₃, of the formula CF₂XCOONa or of the formula CFX₂COONa; or

(g₁) for the preparation of a compound of formula (I) wherein X₁ and X₂ are chlorine or bromine, a compound of formula (IV) is reacted in the presence of a base with a compound of formula



wherein L₃ is a leaving group, preferably chlorine or bromine, and Hal is chlorine or bromine; or

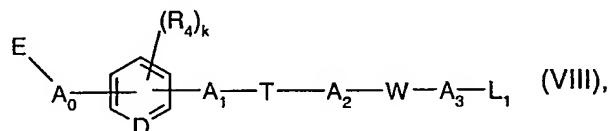
(g₂) for the preparation of a compound of formula (I) wherein X₁ and X₂ are chlorine or bromine, a compound of formula (IV) is reacted in the presence of a base with a compound of formula



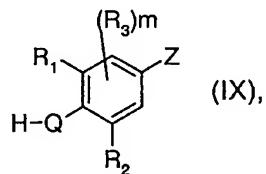
wherein Hal is halogen and X is chlorine or bromine.

The invention relates also to

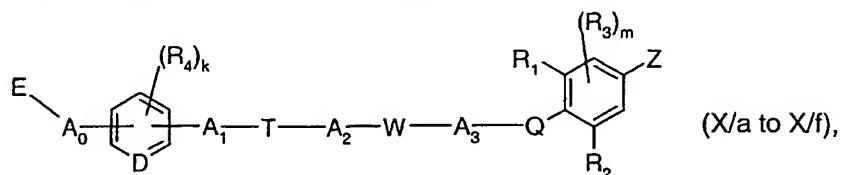
(h) a process for the preparation of a compound of formula (I) as defined under (1) and wherein Q is O, NR₆ or S and R₆ is as defined for formula (I) under (1), wherein a compound of formula



wherein A₀, A₁, A₂, A₃, D, E, W, T, R₄, R₅, R₆ and k are as defined for formula (I) under (1) and L₁ is a leaving group, is reacted in the presence of a base with a compound of formula



wherein R₁, R₂, R₃ and m are as defined for formula (I) under (1), Q is O, NR₆ or S and Z is one of the radicals Z₁ to Z₆ as defined for the above formulae (II) to (VII), and R₆ is as defined for formula (I) under (1), and the resulting compound of formula

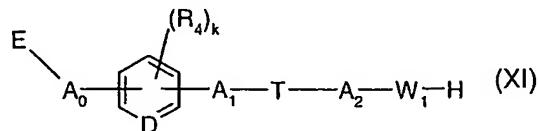


wherein A₀, A₁, A₂, A₃, D, E, W, Q, T, R₁, R₂, R₃, R₄, m and k are as defined for formula (I) under (1) and Z is one of the radicals Z₁ to Z₆ as defined for formulae (II) to (VII) indicated above, is, as necessary, that is to say according to the meaning of the radical Z, reacted further analogously to one or more of process steps (a) to (g).

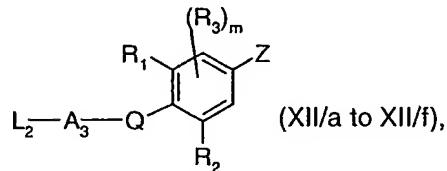
In the compounds of formulae X/a to X/f, Z in compound X/a has the same meanings as Z₁ in the compound of formula (II), and Z in compound X/b has the same meanings as Z₂ as defined for formula (III), and so on.

The invention relates also to

(i₁) a process for the preparation of a compound of formula (I) as defined above wherein W is O, NR₆, S, -O-C(=O)- or -NR₇-C(=O)- and R₆ and R₇ are as defined for formula (I) under (1), wherein a compound of formula

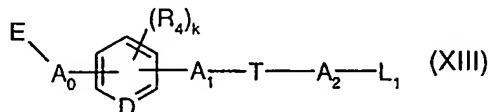


wherein A₀, A₁, A₂, A₃, D, T, R₄ and k are as defined for formula (I) under (1), W₁ is O, NR₆, S or -NR₇- and R₆ is as defined for formula (I) under (1), is reacted with a compound of formula

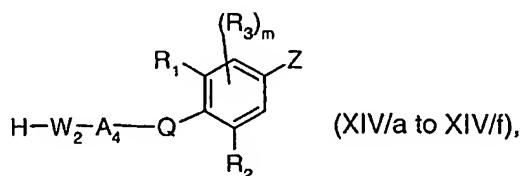


wherein A₃, R₁, R₂, R₃, Q and m are as defined for formula (I) under (1), L₂ is a leaving group or a group Hal-C(=O)- wherein Hal is a halogen atom, preferably chlorine or bromine, and Z is one of the radicals Z₁ to Z₆ as defined in formulae (II) to (VII) indicated above; or

(i₂) for the preparation of a compound of formula (I) as defined above wherein W is O, NR₆, S, -C(=O)-O- or -C(=O)-NR₇- and R₆ and R₇ are as defined for formula (I) under (1), wherein a compound of formula



wherein A₀, A₁, A₂, D, E, T, R₄ and k are as defined for formula (I) under (1) and L₁ is a leaving group or a group -C(=O)-Hal wherein Hal is a halogen atom, preferably chlorine or bromine, is reacted with a compound of formula



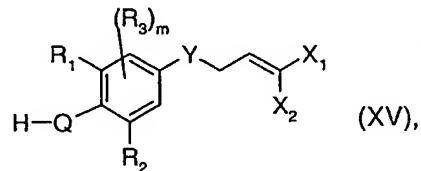
wherein W_2 is O, NR₆, S or NR₇ and R₁, R₂, R₃ and m are as defined for formula (I) under (1),

and a resulting compound of formula (Xa) to (Xf) as defined above is, as necessary, that is to say according to the meaning of the radical Z, reacted further analogously to one or more of process steps (a) to (g).

In the compounds of formulae XII/a to XII/f and XIV/a to XIV/f, the radicals Z are as defined above for the compounds X/a to X/f; that is to say, for example, Z in the compound of formula XII/a has the same meanings as Z₁ in the compound of formula (II), and Z in compound XII/b has the same meanings as Z₂ as defined for formula (III), and so on.

The invention relates also to

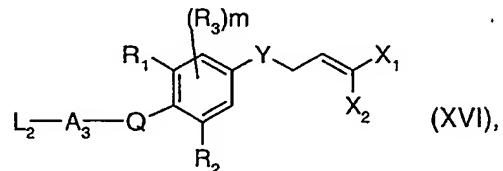
(k) a process for the preparation of a compound of formula (I) as defined above under (1), wherein a compound of formula (VIII) as defined above is reacted in the presence of a base with a compound of formula



wherein R₁, R₂, R₃, Q, X₁, X₂, Y and m are as defined for formula (I) under (1).

The invention relates also to

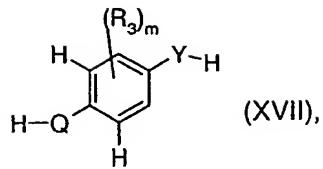
(l) a process for the preparation of a compound of formula (I) as defined above under (1), wherein a compound of formula (XI) as defined above is reacted in a manner analogous to that in process variant (i) with a compound of formula



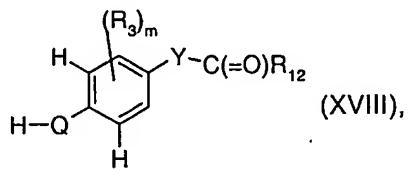
wherein A₃, R₁, R₂, R₃, Q, Y, X₁, X₂ and m are as defined for formula (I) under (1) and L₂ is as defined for formula (XII).

The compounds of formulae (IIIa) and (IIIb) wherein R₁ and R₂ are halogen can be obtained by reacting a compound of formula

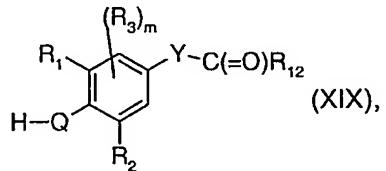
- 15 -



wherein R₃, Q, Y and m are as defined for formula (I) under (1), with a compound of the formula Hal-C(=O)-R₁₃ wherein R₁₃ is as defined above, halogenating the resulting compound of formula



wherein R₃, Q, Y and m are as defined for formula (I) under (1), and further reacting the resulting compound of formula



wherein R₃, Q, Y and m are as defined for formula (I) under (1) and R₁ and R₂ are halogen, analogously to Process (k).

The invention relates also to

(m) a process for the preparation of a compound of formula (II), (III), (IV), (V), (VI), (VII), (VIII), (X), (XI) or (XIII) wherein E is unsubstituted or substituted 1,2,3,4-tetrazol-5-yl, wherein any compound of formula (II), (III), (IV), (V), (VI), (VII), (VIII), (X), (XI) or (XIII) wherein CN is present instead of the heterocyclic radical E is reacted with sodium azide and then substituted further, if desired.

The invention relates also to

(n) a process for the preparation of a compound of formula (II), (III), (IV), (V), (VI), (VII), (VIII), (X), (XI) or (XIII) wherein E is unsubstituted or substituted 1,2,3,4-tetrazol-1-yl, wherein any compound of formula (II), (III), (IV), (V), (VI), (VII), (VIII), (X), (XI) or (XIII), wherein NCS is present instead of the heterocyclic radical E, is reacted with sodium azide and then substituted further, if desired.

The invention relates also to

(o) a process for the preparation of a compound of formula (II), (III), (IV), (V), (VI), (VII), (VIII), (X), (XI) or (XIII) wherein E is unsubstituted or substituted 1,2,4-oxadiazol-3-yl, wherein any compound of formula (II), (III), (IV), (V), (VI), (VII), (VIII), (X), (XI) or (XIII) wherein CN is present instead of the heterocyclic radical E is reacted with hydroxylamine and then with an acid chloride, acid anhydride or orthoformic acid ester, and is then substituted further, if desired.

The invention relates also to

(p) a process for the preparation of a compound of formula (II), (III), (IV), (V), (VI), (VII), (VIII), (X), (XI) or (XIII) wherein E is isoxazol-3-yl, wherein any compound of formula (II), (III), (IV), (V), (VI), (VII), (VIII), (X), (XI) or (XIII), wherein CHO is present instead of the heterocyclic radical E, is reacted with hydroxylamine, then with a halogenating agent and subsequently, in the presence of a base, with an alkyne.

The invention relates also to

(q) a process for the preparation of a compound of formula (II), (III), (IV), (V), (VI), (VII), (VIII), (X), (XI) or (XIII) wherein E is 5-chloro-isoxazol-3-yl, wherein any compound of formula (II), (III), (IV), (V), (VI), (VII), (VIII), (X), (XI) or (XIII), wherein CHO is present instead of the heterocyclic radical E, is reacted with hydroxylamine, then with a halogenating agent and subsequently, in the presence of a base, with 1,1-dichloroethane.

The invention relates also to

(r) a process for the preparation of a compound of formula (II), (III), (IV), (V), (VI), (VII), (VIII), (X), (XI) or (XIII) wherein E is thiazol-2-yl, said thiazole optionally being substituted in the 4-position with alkyl, haloalkyl, hydroxyalkyl, cycloalkyl, cycloalkyl-alkyl, alkenyl, haloalkenyl, alkynyl or haloalkynyl, wherein any compound of formula (II), (III), (IV), (V), (VI), (VII), (VIII), (X), (XI) or (XIII), wherein C(=S)NH₂ is present instead of the heterocyclic radical E, is reacted with a suitable α -haloketone or an α -halocarboxylic acid ester; or

(s) a process for the preparation of a compound of formula (II), (III), (IV), (V), (VI), (VII), (VIII), (X), (XI) or (XIII) wherein E is thiazol-4-yl, said thiazole optionally being substituted in the 2- and in the 4-position independently of each other with alkyl, haloalkyl, hydroxyalkyl, cycloalkyl, cycloalkyl-alkyl, alkenyl, haloalkenyl, alkynyl or haloalkynyl, wherein any compound of formula (II), (III), (IV), (V), (VI), (VII), (VIII), (X), (XI) or (XIII), wherein –

$C(=O)C_1-C_4\text{alkyl}$ is present instead of the heterocyclic radical E, is reacted with an halogenating agent followed by a reaction with a suitable thioamide;

The invention relates also to

(t) a process for the preparation of a compound of formula (II), (III), (IV), (V), (VI), (VII), (VIII), (X), (XI) or (XIII) wherein E is 1,3,4-thiadiazol-2-yl, wherein any compound of formula (II), (III), (IV), (V), (VI), (VII), (VIII), (X), (XI) or (XIII), wherein $B(OH)_2$ is present instead of the heterocyclic radical E, is reacted in the presence of a palladium catalyst with a 2-halo-1,3,4-thiadiazole.

The invention relates also to

(t1) a process for the preparation of a compound of formula (II), (III), (IV), (V), (VI), (VII), (VIII), (X), (XI) or (XIII) wherein E is 1,3,4-thiadiazol-2-yl, wherein any compound of formula (II), (III), (IV), (V), (VI), (VII), (VIII), (X), (XI) or (XIII), wherein halogen or $OS(O_2)CF_3$ is present instead of the heterocyclic radical E, is reacted in the presence of a palladium catalyst with 1,3,4-thiadiazole-2-boronic acid or a 1,3,4-thiadiazole-2-boronic acid ester.

The invention relates also to

(u) a process for the preparation of a compound of formula (II), (III), (IV), (V), (VI), (VII), (VIII), (X), (XI) or (XIII) wherein E is 1,2,4-triazol-3-yl, wherein any compound of formula (II), (III), (IV), (V), (VI), (VII), (VIII), (X), (XI) or (XIII), wherein $-C(=NH)NH_2$ is present instead of the heterocyclic radical E, is reacted in the presence of a base with a carboxylic acid hydrazide.

The reactions described hereinabove and hereinbelow are carried out in a manner known *per se*, for example in the absence or, where appropriate, in the presence of a suitable solvent or diluent or of a mixture thereof, the reactions being carried out, as required, with cooling, at room temperature or with heating, for example in a temperature range of approximately from -80°C to the boiling temperature of the reaction mixture, preferably from approximately -20°C to approximately +150°C, and, if necessary, in a closed vessel, under pressure, under an inert gas atmosphere and/or under anhydrous conditions. Especially advantageous reaction conditions can be found in the Examples.

A leaving group, for example the leaving groups L₁ and L₂ defined above, or a counter-ion is to be understood hereinabove and hereinbelow as being any removable group that customarily comes into consideration for chemical reactions, such as are known to the person skilled in the art; especially OH, halogens, such as fluorine, chlorine, bromine, iodine,

-O-Si(C₁-C₈alkyl)₃, -O-aryl, -S-(C₁-C₈alkyl), -S-aryl, -O-S(=O)₂U, -S(=O)U or -S(=O)₂U, wherein U is unsubstituted or substituted C₁-C₈alkyl, C₂-C₈alkenyl, C₂-C₈alkynyl, unsubstituted or substituted aryl or unsubstituted or substituted benzyl. Especially preferred as leaving group are chlorine and bromine, mesylate, triflate, tosylate, especially chlorine; and chloride and bromide, especially chloride.

Process (a): The reaction is carried out in acetic acid or a halogenated hydrocarbon, such as dichloromethane, at temperatures of from -20°C to 100°C, preferably at from 20°C to 50°C. As oxidising agents there are used, for example, hydrogen peroxide, a peracid, such as peracetic acid, trifluoroperacetic acid, 3-chloroperbenzoic acid or a mixture thereof, such as sodium perborate in acetic acid.

Process (b): The reaction is preferably carried out in an alcohol, such as methanol, ethanol or an alcohol/water mixture, in the presence of an inorganic base, such as NaOH or KOH, and at temperatures of from 0°C to 150°C, preferably from 20°C to 80°C. Alternatively aminolysis with a primary amine, such as n-butylamine, can be carried out in a hydrocarbon, such as toluene or benzene, at temperatures of from 0°C to 150°C, preferably at from 20°C to 80°C.

Process (c): Depending upon the nature of the benzyl substituent to be removed, the reaction can be carried out, for example, under a hydrogen atmosphere, at a pressure of from 1 to 150 bar, especially at from 1 to 20 bar, and with the addition of a catalyst, such as palladium/carbon, in an alcohol or ether. The preferred reaction temperature is from 0°C to 120°C, especially from 20°C to 80°C.

Processes (d) and (g): The reactions are preferably carried out in the presence of a base, such as potassium or sodium carbonate, in acetone or dimethylformamide, at temperatures of from 0°C to 150°C, preferably from 20°C to 80°C. If necessary, catalytic amounts of potassium iodide or sodium iodide, or phase transfer catalysts, such as crown ethers or quaternary ammonium salts, are added.

Process (e): The reaction is preferably carried out in acetone, dichloromethane, acetic acid, or especially in water, optionally with the addition of a mineral acid, at temperatures of from 0°C to 120°C, preferably at from 20°C to 50°C. For complete cleavage of the acetal it is preferable to add a strong mineral acid, for example hydrochloric acid, sulfuric acid or 4-toluenesulfonic acid.

Process (f): For the preparation of the difluoro-, dichloro-, dibromo-, chlorofluoro- and bromofluoro-vinyl compounds, reaction with CCl_4 , CBr_4 , CF_2X_2 , CFX_3 , $\text{CF}_2\text{XC}(=\text{O})\text{ONa}$ or $\text{CFX}_2\text{C}(=\text{O})\text{ONa}$ wherein X is bromine or chlorine is carried out in the presence of a trialkyl- or triaryl-phosphine, optionally with the addition of powdered zinc. The reaction is carried out in an inert solvent such as, for example, benzene or toluene, or an ether, such as diethyl ether, diisopropyl ether, dioxane or tetrahydrofuran, at temperatures of from 0°C to 150°C, preferably at from 20°C to 80°C.

For the preparation of the dichlorovinyl compounds it is also possible for the process to be carried out in dimethylformamide, benzene, toluene, or in an ether, at temperatures of from 0°C to 120°C, preferably from 20°C to 80°C, and in the presence of trichloroacetic acid/sodium trichloroacetate, then by addition of acetic anhydride, optionally with the addition of base, for example triethylamine, and finally by addition of zinc and acetic acid.

Processes (h) and (k): The reactions are preferably carried out in an ether, dimethylformamide, dimethylacetamide or N-methylpyrrolidone, at temperatures of from 0°C to 150°C, preferably at from 20°C to 80°C, with the addition of a base, such as potassium or sodium carbonate. Alternatively a coupling reagent, for example azodicarboxylic acid diethyl or diisopropyl ester and triphenylphosphine, can be used.

Processes (i) and (l):

Where L_2 is a group $\text{Hal}-\text{C}(=\text{O})-$, the process can be carried out in an inert solvent, such as in an ether or in toluene, at from 0°C to 80°C, and in the presence of a suitable base, for example a trialkylamine.

In the other cases the reaction is carried out in an ether, in an amide, such as dimethylformamide or N-methylpyrrolidone, and at from 0°C to 150°C. Sodium hydride, for example, can be used as base.

Process (m): There are suitable as solvents, for example, toluene, benzene, ethanol, propanol, isopropanol, methoxyethanol, ethoxyethanol or dimethylformamide. The reaction is carried out at from 20 to 200°C, preferably at from 80 to 150°C, where appropriate with the addition of lithium chloride, ammonium chloride or triethylammonium chloride.

The further reaction is carried out, for example, in dimethylformamide, dimethyl sulfoxide, tetrahydrofuran, acetonitrile or toluene, in the presence of a base, for example potassium or sodium carbonate.

Process (n): The reaction is carried out, for example, in toluene, benzene, dioxane, tetrahydrofuran, dimethylformamide or N-methylpyrrolidone, at from 0 to 130°C, preferably at from 20 to 80°C.

Process (o): The reaction is carried out with hydroxylamine hydrochloride in the presence of a base, such as triethylamine, for example in ethanol, isopropanol, dimethylformamide or N-methylpyrrolidone, at from 0 to 120°C, preferably at from 20 to 80°C.

The further reaction is carried out, for example, in toluene, benzene, THF, dioxane or dichloromethane, at from 0 to 120°C in the presence of a base, such as triethylamine or pyridine.

Process (p): The reaction with hydroxylamine hydrochloride is carried out in the presence of a base, such as triethylamine. The halogenation of the resulting oxime is carried out, for example, using sodium hypochlorite or tert-butyl hypochlorite in carbon tetrachloride, chloroform or dichloromethane.

The further reaction with an alkyne is carried out in the presence of a base, such as triethylamine or pyridine, at from -20 to 120°C, preferably at from 0 to 80°C.

Process (q): The process is carried out analogously to procedure (p). Instead of an alkyne, 1,1-dichloroethane is used.

Processes (r) and (s): The reaction is carried out, for example, in ethanol, propanol or dimethylformamide, at from 0 to 120°C, preferably at from 20 to 80°C.

Process (t): The Reaction is for example carried out in toluene, benzene, an ether, such as dimethoxyethane, in the presence of a base, for example aqueous sodium carbonate solution, and a palladium catalyst, such as palladium tetrakis(triphenylphosphine), at temperatures of from 0 to 150°C, preferably at from 20 to 100°C.

Process (u): The reaction of the amidine is carried out, for example, in benzene, toluene, diethyl ether, diisopropyl ether, tert-butyl methyl ether, in the presence of a base, for example potassium or sodium hydroxide, at from 0 to 150°C, preferably at from 20 to 100°C.

It will be understood that in the above-described Processes (m) to (u), in which a compound of formula (II), (III), (IV), (V), (VI), (VII), (VIII), (X), (XI) or (XIII) carrying a substituent, such as CN, -NCS, B(OH)₂, -OS(O₂)CF₃, -C(=O)O-alkyl, -C(=NH)NH₂, I, Br or CHO instead of the heterocyclic radical E is processed further, further functional groups

likewise carried by those intermediate compounds must under certain circumstances be protected by protecting groups. Such protecting groups are introduced according to the nature of the substituent in question. They include any removable group that customarily comes into consideration for chemical reactions, such as are known to the person skilled in the art. For example, aldehyde and keto groups are protected in the form of acetals or ketals, acids in the form of their esters, hydroxy groups in the form of ethers or esters, and phenolic hydroxy groups in the form of benzyl ethers or methyl ethers.

Compounds of formula (I) obtainable in accordance with the process or by other means can be converted into other compounds of formula (I) in a manner known *per se* by replacement of one or more substituents in the starting compound of formula (I) in customary manner by another (other) substituent(s) according to the invention.

In the case of such replacement, depending upon the choice of reaction conditions and starting materials suitable therefor, it is possible for only one substituent to be replaced by another substituent according to the invention in a reaction step or for a plurality of substituents to be replaced by other substituents according to the invention in the same reaction step.

Salts of compounds of formula (I) can be prepared in a manner known *per se*. For example, salts of compounds of formula (I) with bases are obtained by treatment of the free compounds with a suitable base or with a suitable ion exchange reagent.

Salts of compounds of formula (I) can be converted into the free compounds of formula (I) in customary manner, for example by treatment with a suitable acid or with a suitable ion exchange reagent.

Salts of compounds of formula (I) can be converted in a manner known *per se* into other salts of a compound of formula (I).

The compounds of formula (I), in free form or in salt form, may be in the form of one of the possible isomers or in the form of a mixture thereof, for example, depending upon the number of asymmetric carbon atoms occurring in the molecule and their absolute and relative configuration, and/or depending upon the configuration of non-aromatic double bonds occurring in the molecule, in the form of pure isomers, such as antipodes and/or diastereoisomers, or in the form of mixtures of isomers, such as mixtures of enantiomers, for example racemates, mixtures of diastereoisomers or mixtures of racemates. The invention relates both to the pure isomers and to all possible mixtures of isomers and is to be

interpreted as such hereinbefore and hereinafter, even if stereochemical details are not mentioned specifically in every case.

Mixtures of diastereoisomers, mixtures of racemates and mixtures of double bond isomers of compounds of formula (I), in free form or in salt form, which may be obtained in accordance with the process – depending upon the starting materials and procedures chosen – or by some other method, can be separated into the pure diastereoisomers or racemates in known manner on the basis of the physico-chemical differences between the constituents, for example by means of fractional crystallisation, distillation and/or chromatography.

Mixtures of enantiomers, such as racemates, that are obtainable in a corresponding manner can be resolved into the enantiomers by known methods, for example by recrystallisation from an optically active solvent, by chromatography on chiral adsorbents, for example high pressure liquid chromatography (HPLC) on acetylcellulose, with the aid of suitable microorganisms, by cleavage with specific, immobilised enzymes, *via* the formation of inclusion compounds, for example using chiral crown ethers, only one enantiomer being complexed, or by conversion into diastereoisomeric salts and separation of the mixture of diastereoisomers so obtained, for example on the basis of their different solubilities by fractional crystallisation, into the diastereoisomers, from which the desired enantiomer can be freed by the action of suitable agents.

Apart from by separation of corresponding mixtures of isomers, pure diastereoisomers or enantiomers can be obtained according to the invention also by generally known methods of diastereoselective or enantioselective synthesis, for example by carrying out the process according to the invention using starting materials having correspondingly suitable stereochemistry.

In each case it is advantageous to isolate or synthesise the biologically more active isomer, e.g. enantiomer or diastereoisomer, or isomeric mixture, e.g. enantiomeric mixture or diastereoisomeric mixture, where the individual components have different biological activity.

The compounds of formula (I), in free form or in salt form, can also be obtained in the form of their hydrates and/or may include other solvents, for example solvents which may have been used for the crystallisation of compounds in solid form.

The invention relates to all those embodiments of the process according to which a compound obtainable as starting material or intermediate at any stage of the process is used

as starting material and some or all of the remaining steps are carried out or a starting material is used in the form of a derivative or salt and/or its racemates or antipodes or, especially, is formed under the reaction conditions.

In the process of the present invention it is preferable to use those starting materials and intermediates, in each case in free form or in salt form, which result in the compounds of formula (I) and their salts described at the beginning as being especially valuable.

The invention relates especially to the preparation processes described in Examples P1 to P11.

The invention relates also to the intermediates of formulae (II) to (XIX), where novel, and to the corresponding compounds wherein CN, NCS, C(H)O, B(OH)₂, -C(H)=NOH, -C(Cl)=NOH, -C(=NH)NH₂ or -C(=S)NH₂ is present instead of the heterocyclic radical E, and, where applicable, to their possible E/Z isomers, E/Z isomeric mixtures and/or tautomers, in each case in free form or in salt form. The preferences applying to such compounds are the same as those for the compounds of formula (I).

In the area of pest control, the compounds of formula (I) according to the invention are active ingredients exhibiting valuable preventive and/or curative activity with a very advantageous biocidal spectrum and a very broad spectrum, even at low rates of concentration, while being well tolerated by warm-blooded animals, fish and plants. They are, surprisingly, equally suitable for controlling both plant pests and ecto- and endo-parasites in humans and more especially in productive livestock, domestic animals and pets. They are effective against all or individual development stages of normally sensitive animal pests, but also of resistant animal pests, such as insects and representatives of the order Acarina, nematodes, cestodes and trematodes, while at the same time protecting useful organisms. The insecticidal or acaricidal activity of the active ingredients according to the invention may manifest itself directly, i.e. in the mortality of the pests, which occurs immediately or only after some time, for example during moulting, or indirectly, for example in reduced oviposition and/or hatching rate, good activity corresponding to a mortality of at least 50 to 60 %.

Successful control within the scope of the subject of the invention is possible, in particular, of pests from the orders Lepidoptera, Coleoptera, Orthoptera, Isoptera, Psocoptera, Anoplura, Mallophaga, Thysanoptera, Heteroptera, Homoptera, Hymenoptera, Diptera, Siphonaptera, Thysanura and Acarina, mainly Acarina, Diptera, Thysanoptera, Lepidoptera and Coleoptera. Very especially good control is possible of the following pests:

Abagrotis spp., Abraxas spp., Acantholeucania spp., Acanthoplusia spp., Acarus spp.,
Acarus siro, Aceria spp., Aceria sheldoni, Acleris spp., Acoloithus spp., Acompsia spp.,
Acossus spp., Acria spp., Acrobasis spp., Acrocercops spp., Acrolepia spp., Acrolepiopsis
spp., Acronicta spp., Acropolitis spp., Actebia spp., Aculus spp., Aculus schlechtendali,
Adoxophyes spp., Adoxophyes reticulana, Aedes spp., Aegeria spp., Aethes spp., Agapeta
spp., Agonopterix spp., Agriopis spp., Agriotes spp., Agriphila spp., Agrochola spp., Agro-
perina spp., Alabama spp., Alabama argillaceae, Agrotis spp., Albuna spp., Alcathoe spp.,
Alcis spp., Aleimma spp., Aletia spp., Aleurothrixus spp., Aleurothrixus floccosus, Aleyrodes
spp., Aleyrodes brassicae, Allophyes spp., Alsophila spp., Amata spp., Amathes spp.,
Amblyomma spp., Amblyptilia spp., Ammoconia spp., Amorbia spp., Amphion spp., Amphi-
poea spp., Amphyipyra spp., Amyelois spp., Anacamptodes spp., Anagrapha spp., Anarsia
spp., Anatrychytis spp., Anavitrinella spp., Ancylis spp., Andropolia spp., Anhimella spp.,
Antheraea spp., Antherigona spp., Antherigona saccata, Anthonomus spp., Anthonomus
grandis, Anticarsia spp., Anticarsia gemmatalis, Aonidiella spp., Apamea spp., Aphania spp.,
Aphelia spp., Aphididae, Aphis spp., Apotomis spp., Aproaerema spp., Archippus spp.,
Archips spp., Acromyrmex, Arctia spp., Argas spp., Argolamprotes spp., Argyresthia spp.,
Argyrogramma spp., Argyroploce spp., Argyrotaenia spp., Arotrophora spp., Ascotis spp.,
Aspidiotus spp., Aspilapteryx spp., Asthenopterycha spp., Aterpia spp., Athetis spp., Atomaria
spp., Atomaria linearis, Atta spp., Atypha spp., Autographa spp., Axylia spp., Bactra spp.,
Barbara spp., Batrachedra spp., Battaristis spp., Bembecia spp., Bernisia spp., Bernisia
tabaci, Bibio spp., Bibio hortulanis, Bisigna spp., Blastesthia spp., Blatta spp., Blatella spp.,
Blepharosis spp., Bleptina spp., Boarmia spp., Bombyx spp., Bomolocha spp., Boophilus
spp., Brachmia spp., Bradina spp., Brevipalpus spp., Brithys spp., Bryobia spp., Bryobia
praetiosa, Bryotropha spp., Bupalus spp., Busseola spp., Busseola fusca, Cabera spp.,
Cacoecimorpha spp., Cadra spp., Cadra cautella, Caenurgina spp., Calipitrimerus spp.,
Callierges spp., Callophpora spp., Callophora erythrocephala, Calophasia spp., Caloptilia
spp., Calybites spp., Capnoptycha spp., Capua spp., Caradrina spp., Caripeta spp., Car-
menta spp., Carposina spp., Carposina nipponensis, Catamacta spp., Catelaphris spp.,
Catoptria spp., Caustoloma spp., Celaena spp., Celypha spp., Cenopis spp., Cephus spp.,
Ceramica spp., Cerapteryx spp., Ceratitis spp., Ceratophyllus spp., Ceroplasster spp., Chaeto-
cnema spp., Chaetocnema tibialis, Chamaesphecia spp., Charanvca spp., Cheimophila spp.,
Chersotis spp., Chiasmia spp., Chilo spp., Chionodes spp., Chorioptes spp., Choristoneura
spp., Chrysaspidea spp., Chrysodeixis spp., Chrysomya spp., Chrysomphalus spp., Chry-
somphalus dictyospermi, Chrysomphalus aonidium, Chrysoteuchia spp., Cilix spp., Cimex

spp., *Clytia* spp., *Clytia ambiguella*, *Clepsis* spp., *Cnaemidophorus* spp., *Cnaphalocrosis* spp., *Cnephasia* spp., *Coccus* spp., *Coccus hesperidum*, *Cochylis* spp., *Coleophora* spp., *Colotois* spp., *Commophila* spp., *Conistra* spp., *Conopomorpha* spp., *Corcyra* spp., *Cornutiplusia* spp., *Cosmia* spp., *Cosmopolites* spp., *Cosmopterix* spp., *Cossus* spp., *Costaeonvexa* spp., *Crambus* spp., *Creatonotos* spp., *Crocidolomia* spp., *Crocidolomia binotalis*, *Croesia* spp., *Crymodes* spp., *Cryptaspasma* spp., *Cryptoblabes* spp., *Cryptocala* spp., *Cryptophlebia* spp., *Cryptophlebia leucotreta*, *Cryptoptila* spp., *Ctenopseustis* spp., *Cucullia* spp., *Curculio* spp., *Culex* spp., *Cuterebra* spp., *Cydia* spp., *Cydia pomonella*, *Cymbalophora* spp., *Dactylethora* spp., *Dacus* spp., *Dadica* spp., *Damalinea* spp., *Dasychira* spp., *Decadrachis* spp., *Decodes* spp., *Deilephila* spp., *Deltodes* spp., *Dendrolimus* spp., *Depressaria* spp., *Dermestes* spp., *Dermanyssus* spp., *Dermanyssus gallinae*, *Diabrotica* spp., *Diachrysia* spp., *Diaphania* spp., *Diarsia* spp., *Diasemia* spp., *Diatraea* spp., *Diceratura* spp., *Dichomeris* spp., *Dichrocrocis* spp., *Dichrorampha* spp., *Dicycla* spp., *Dioryctria* spp., *Diparopsis* spp., *Diparopsis castanea*, *Dipleurina* spp., *Diprion* spp., *Diprionidae*, *Discestra* spp., *Distantiella* spp., *Distantiella theobroma*, *Ditula* spp., *Diurnea* spp., *Doratopteryx* spp., *Drepana* spp., *Drosophila* spp., *Drosophila melanogaster*, *Dysauxes* spp., *Dysdercus* spp., *Dysstroma* spp., *Eana* spp., *Earias* spp., *Ecclitica* spp., *Ecdytolopha* spp., *Ecpyrrhorhoe* spp., *Ectomyelois* spp., *Eetropis* spp., *Egira* spp., *Elasmopalpus* spp., *Emmelia* spp., *mpoasca* spp., *Empyreuma* spp., *Enargia* spp., *Enarmonia* spp., *Endopiza* spp., *Endothenia* spp., *Endotricha* spp., *Eoreuma* spp., *Eotetranychus* spp., *Eotetranychus carpini*, *Epagoge* spp., *Epelis* spp., *Ephestia* spp., *Ephestiodes* spp., *Epiblema* spp., *Epiehoristodes* spp., *Epinotia* spp., *Epiphyas* spp., *Epiblema* spp., *Epipsestis* spp., *Epirrhoe* spp., *Episimus* spp., *Epitymbia* spp., *Epilachna* spp., *Erannis* spp., *Erastria* spp., *Eremnus* spp., *Ereunetis* spp., *Eriophyes* spp., *Eriosoma* spp., *Eriosoma lanigerum*, *Erythroneura* spp., *Estigmene* spp., *Ethmia* spp., *Etiella* spp., *Euagrotis* spp., *Eucosma* spp., *Euehlaena* spp., *Euelidia* spp., *Eueosma* spp., *Euchistus* spp., *Eucosmomorpha* spp., *Eudonia* spp., *Eufidonia* spp., *Euhyonomeutoides* spp., *Eulepidotes* spp., *Eulia* spp., *Eulithis* spp., *Eupithecia* spp., *Euplexia* spp., *Eupoecilia* spp., *Eupoecilia ambiguella*, *Euproctis* spp., *Eupsilia* spp., *Eurhodope* spp., *Eurois* spp., *Eurygaster* spp., *Eurythmia* spp., *Eustrotia* spp., *Euxoa* spp., *Euzophera* spp., *Evergestis* spp., *Evippe* spp., *Exarterma* spp., *Fannia* spp., *Faronta* spp., *Feltia* spp., *Filatima* spp., *Fishia* spp., *Frankliniella* spp., *Fumibotys* spp., *Gaesota* spp., *Gasgardia* spp., *Gastrophilus* spp., *Gelechia* spp., *Gilpinia* spp., *Gilpinia polytoma*, *Glossina* spp., *Glyptipterix* spp., *Glyphodes* spp., *Gnorimoschemini* spp., *Gonodonta* spp., *Gortyna* spp., *Gracillaria* spp., *Graphania* spp., *Grapholita* spp., *Grapholitha* spp., *Gravitarmata* spp., *Gretchenia* spp.,

Griselda spp., Gryllotalpa spp., Gynaephora spp., Gypsonoma spp., Hada spp., Haematoxinus spp., Halisidota spp., Harpipteryx spp., Harrisina spp., Hedyia spp., Helicoverpa spp., Heliophobus spp., Heliothis spp., Hellula spp., Helotropa spp., Hemaris spp., Hercinothrips spp., Herculia spp., Hermonassa spp., Heterogenea spp., Holomelina spp., Homadaula spp., Homoeosoma spp., Homoglaea spp., Homohadena spp., Homona spp., Homonopsis spp., Hoplocampa spp., Hoplodrina spp., Hoshinoa spp., Hxalomma spp., Hydraecia spp., Hydriomena spp., Hyles spp., Hyloicus spp., Hypagyrtis spp., Hypatima spp., Hyphantria spp., Hyphantria cunea, Hypocala spp., Hypocoena spp., Hypodema spp., Hyppobosca spp., Hypsipyla spp., Hyssia spp., Hysterosia spp., Idaea spp., Idia spp., Ipimorpha spp., Isia spp., Isochorista spp., Isophrictis spp., Isopolia spp., Isotrias spp., Ixodes spp., Itame spp., Jodia spp., Jodis spp., Kawabea spp., Keiferia spp., Keiferia lycopersicella, Labdia spp., Laciniopolia spp., Lambdina spp., Lamprothritpa spp., Laodelphax spp., Lasius spp., Laspeyresia spp., Leptinotarsa spp., Leptinotarsa decemlineata, Leptocoris spp., Leptostales spp., Lecanium spp., Lecanium comi, Lepidosaphes spp., Lepisma spp.; Lepisma saccharina, Lesmone spp., Leucania spp., Leucinodes spp., Leucophaea spp., Leucophaea maderae, Leucoptera spp., Leucoptera scitella, Linognathus spp., Liposcelis spp., Lissorhoptrus spp., Lithacodia spp., Lithocolletis spp., Lithomoia spp., Lithophane spp., Lixodessa spp., Lobesia spp., Lobesia botrana, Lobophora spp., Locusta spp., Lomanaltes spp., Lomographa spp., Loxagrotis spp., Loxostege spp., Lucilia spp., Lymantria spp., Lymnaecia spp., Lyonetia spp., Lyriomyza spp., Macdonnoughia spp., Macrauzata spp., Macro noctua spp., Macrosiphus spp., Malacosoma spp., Maliarpha spp., Mamestra spp., Mamestra brassicae, Manduca spp., Manduca sexta, Marasmia spp., Margaritiæ spp., Matratinea spp., Matsumuraes spp., Melanagromyza spp., Melipotes spp., Melissopus spp., Melittia spp., Melolontha spp., Meristis spp., Meritastis spp., Merophyas spp., Mesapamea spp., Mesogona spp., Mesoleuca spp., Metanema spp., Metendothenia spp., Metzneria spp., Micardia spp., Microcorses spp., Microleon spp., Mnesictena spp., Mocis spp., Monima spp., Monochroa spp., Monomorium spp., Monomorium pharaonis, Monopsis spp., Morrisonia spp., Musca spp., Mutuuraia spp., Myelois spp., Mythimna spp., Myzus spp., Naranga spp., Nedra spp., Nemopagon spp., Neodiprion spp., Neosphaleroptera spp., Nephelodes spp., Nephrotettix spp., Nezara spp., Nilaparvata spp., Niphonympha spp., Nippoptilia spp., Noctua spp., Nola spp., Notocelia spp., Notodontia spp., Nudaurelia spp., Ochropleura spp., Ocnerostoma spp., Oestrus spp., Olethreutes spp., Oligia spp., Olindia spp., Olygonychus spp., Olygonychus gallinae, Oncocnemis spp., Operophtera spp., Ophisma spp., Opogona spp., Oraesia spp., Orniodoros spp., Orgyia spp., Orias spp., Orseolia spp., Orthodes spp., Orthogonia spp.,

Orthosia spp., Oryzaephilus spp., Oscinella spp., Oscinella frit, Osminia spp., Ostrinia spp., Ostrinia nubilalis, Otiorhynchus spp., Ourapteryx spp., Pachetra spp., Pachysphinx spp., Pagyda spp., Paleacrita spp., Paliga spp., Palthis spp., Pammene spp., Pandemis spp., Panemera spp., Panolis spp., Panolis flammea, Panonychus spp., Parargyresthia spp., Paradiarsia spp., Paralobesia spp., Paranthrene spp., Parapandemis spp., Parapediasia spp., Parastichtis spp., Parasyndemis spp., Paratoria spp., Pareromeme spp., Pectinophora spp., Pectinophora gossypiella, Pediculus spp., Pegomyia spp., Pegomyia hyoscyami, Pelochrista spp., Pennisetia spp., Penstemonia spp., Pemphigus spp., Peribatodes spp., Peridroma spp., Perileucoptera spp., Periplaneta spp., Perizoma spp., Petrova spp., Pexicopia spp., Phalonia spp., Phalonidia spp., Phaneta spp., Phlyctaenia spp., Phlyctinus spp., Phobia spp., Phragmatobia spp., Phricanthes spp., Phthorimaea spp., Phthorimaea operculella, Phylloconistis spp., Phyllocopruta spp., Phyllocopruta oleivora, Phyllonorycter spp., Phyllophila spp., Phylloxera spp., Pieris spp., Pieris rapae, Piesma spp., Planococcus spp., Planotortrix spp., Platyedra spp., Platynota spp., Platypilia spp., Platysenta spp., Plodia spp., Plusia spp., Plutella spp., Plutella xylostella, Pododesia spp., Polia spp., Popillia spp., Polymixis spp., Polyphagotarsonemus spp., Polyphagotarsonemus latus, Prays spp., Prionoxystus spp., Probola spp., Proceras spp., Prochoerodes spp., Proeulia spp., Proschistis spp., Proselena spp., Proserpinus spp., Protagrotis spp., Proteoteras spp., Protobathra spp., Protoschinia spp., Pselnophorus spp., Pseudaletia spp., Pseudanthonomus spp., Pseudaternelia spp., Pseudaulacaspis spp., Pseudexentera spp., Pseudococcus spp., Pseudohermenias spp., Pseudoplusia spp., Psoroptes spp., Psylla spp., Psylliodes spp., Pterophorus spp., Ptycholoma spp., Pulvinaria spp., Pulvinaria aethiopica, Pyralis spp., Pyrausta spp., Pyrgotis spp., Pyreferra spp., Pyrrharctia spp., Quadraspidiotus spp., Rancora spp., Raphia spp., Reticulitermes spp., Retinia spp., Rhagoletis spp., Rhagoletis pomonella, Rhipicephalus spp., Rhizoglyphus spp., Rhizopertha spp., Rhodnius spp., Rhopalosiphum spp., Rhopobota spp., Rhyacia spp., Rhyacionia spp., Rhynchopacha spp., Rhynchosthenes spp., Rivula spp., Rondotia spp., Rusidrina spp., Rynchaglaea spp., Sabulodes spp., Sahlbergella spp., Sahlbergella singularis, Saissetia spp., Samia spp., Sannina spp., Sanninoidea spp., Saphoideus spp., Sarcoptes spp., Sathrobrota spp., Scarabeidae, Sceloides spp., Schinia spp., Schistocerca spp., Schizaphis spp., Schizura spp., Schreckensteinia spp., Sciara spp., Scirpophaga spp., Scirthrips auranti, Scoparia spp., Scopula spp., Scotia spp., Scotinophara spp., Scotogramma spp., Scrobipalpa spp., Scrobipalopsis spp., Semiothisa spp., Sereda spp., Sesamia spp., Sesia spp., Sicya spp., Sideridis spp., Simyra spp., Sineugrapha spp., Sitochroa spp., Sitobion spp., Sitophilus spp., Sitotroga spp., Solen-

opsis spp., Smerinthus spp., Sophronia spp., Spaelotis spp., Spargaloma spp., Sparganothis spp., Spatialistis spp., Sperchia spp., Sphecia spp., Sphinx spp., Spilonota spp., Spodoptera spp., Spodoptera littoralis, Stagmatophora spp., Staphylinochrous spp., Stathmopoda spp., Stenodes spp., Sterrha spp., Stomoxys spp., Strophedra spp., Sunira spp., Sutyna spp., Swammerdaria spp., Syllomatia spp., Sympistis spp., Synanthesdon spp., Synaxis spp., Syncopacma spp., Syndemis spp., Syngrapha spp., Synthomeida spp., Tabanus spp., Taeniarchis spp., Taeniothrips spp., Tannia spp., Tarsonemus spp., Tegulifera spp., Tehama spp., Teleiodes spp., Telorta spp., Tenebrio spp., Tephritis spp., Teratoglaea spp., Terricula spp., Tethea spp., Tetranychus spp., Thalpophila spp., Thaumetopoea spp., Thiodia spp., Thrips spp., Thrips palmi, Thrips tabaci, Thyridopteryx spp., Thyris spp., Tineola spp., Tipula spp., Tortricidia spp., Tortrix spp., Trachea spp., Trialeurodes spp., Trialeurodes vaporariorum, Triatoma spp., Triaxomera spp., Tribolium spp., Tricodectes spp., Trichoplusia spp., Trichoplusia ni, Trichoptilus spp., Trioza spp., Trioza erytreae, Triphaenia spp., Triphosa spp., Trogoderma spp., Tyria spp., Udea spp., Unaspis spp., Unaspis citri, Utetheisa spp., Valeriodes spp., Vespa spp., Vespamima spp., Vitacea spp., Vitula spp., Witlesia spp., Xanthia spp., Xanthorhoe spp., Xanthotype spp., Xenomicta spp., Xenopsylla spp., Xenopsylla cheopsis, Xestia spp., Xylena spp., Xylomyges spp., Xyrosaris spp., Yponomeuta spp., Ypsolopha spp., Zale spp., Zanclognathus spp., Zeiraphera spp., Zenodoxus spp., Zeuzera spp., Zygaena spp.,

It is also possible to control pests of the class Nematoda using the compounds according to the invention. Such pests include, for example,

root knot nematodes, cyst-forming nematodes and also stem and leaf nematodes; especially of *Heterodera* spp., e.g. *Heterodera schachtii*, *Heterodora avenae* and *Heterodora trifolii*; *Globodera* spp., e.g. *Globodera rostochiensis*; *Meloidogyne* spp., e.g. *Meloidogyne incognita* and *Meloidogyne javanica*; *Radopholus* spp., e.g. *Radopholus similis*; *Pratylenchus*, e.g. *Pratylenchus neglectans* and *Pratylenchus penetrans*; *Tylenchulus*, e.g. *Tylenchulus semipenetrans*; *Longidorus*, *Trichodorus*, *Xiphinema*, *Ditylenchus*, *Apheenchoides* and *Anguina*; especially *Meloidogyne*, e.g. *Meloidogyne incognita*, and *Heterodera*, e.g. *Heterodera glycines*.

An especially important aspect of the present invention is the use of the compounds of formula (I) according to the invention in the protection of plants against parasitic feeding pests.

The action of the compounds according to the invention and the compositions com-

prising them against animal pests can be significantly broadened and adapted to the given circumstances by the addition of other insecticides, acaricides or nematicides. Suitable additives include, for example, representatives of the following classes of active ingredient: organophosphorus compounds, nitrophenols and derivatives, formamidines, ureas, carbamates, pyrethroids, chlorinated hydrocarbons, neonicotinoids and *Bacillus thuringiensis* preparations.

Examples of especially suitable mixing partners include: azamethiphos; chlorgenvinphos; cypermethrin, cypermethrin high-cis; cyromazine; diafenthiuron; diazinon; dichlorvos; dicrotophos; dicyclanil; fenoxy carb; fluazuron; furathiocarb; isazofos; iodfenphos; kinoprene; lufenuron; methacryphon; methidathion; monocrotophos; phosphamidon; profenofos; diofenolan; a compound obtainable from the *Bacillus thuringiensis* strain GC91 or from strain NCTC11821; pymetrozine; bromopropylate; methoprene; disulfoton; quinalphos; tau-fluvalinate; thiocyclam; thiometon; aldicarb; azinphos-methyl; benfuracarb; bifenthrin; buprofezin; carbofuran; dibutylaminothio; cartap; chlorfluazuron; chlorpyrifos; clothianidin; cyfluthrin; lambda-cyhalothrin; alpha-cypermethrin; zeta-cypermethrin; deltamethrin; diflubenzuron; endosulfan; ethiofencarb; fenitrothion; fenobucarb; fenvalerate; formothion; methiocarb; heptenophos; imidacloprid; isoprocarb; methamidophos; methomyl; mevinphos; parathion; parathion-methyl; phosalone; pirimicarb; propoxur; teflubenzuron; terbufos; triazamate; fenobucarb; tebufenozone; fipronil; beta-cyfluthrin; silafluofen; fenpyroximate; pyridaben; fenazaquin; pyriproxyfen; pyrimidifen; nitenpyram; acetamiprid; emamectin; emamectin-benzoate; spinosad; a plant extract that is active against insects; a preparation that comprises nematodes and is active against insects; a preparation obtainable from *Bacillus subtilis*; a preparation that comprises fungi and is active against insects; a preparation that comprises viruses and is active against insects; chlorgafenapyr; acephate; acrinathrin; alanycarb; alphamethrin; amitraz; AZ 60541; azinphos A; azinphos M; azocyclotin; bendiocarb; bensultap; beta-cyfluthrin; BPMC; brofenprox; bromophos A; bufencarb; butocarboxin; butylpyridaben; cadusafos; carbaryl; carbophenothion; chloethocarb; chlorethoxyfos; chlormephos; cis-resmethrin; clocythrin; clofentezine; cyanophos; cycloprothrin; cyhexatin; demeton M; demeton S; demeton-S-methyl; dichlofenthion; dicliphos; diethion; dimethoate; dimethylvinphos; dioxathion; edifenphos; esfenvalerate; ethion; ethofenprox; ethoprophos; etrimphos; fenamiphos; fenbutatin oxide; fenothiocarb; fenpropothrin; fenpyrad; fenthion; fluazinam; flucycloxuron; flucythrinate; flufenoxuron; flufenprox; fonophos; fosthiazate; fubfenprox; HCH; hexaflumuron; hexythiazox; IKI-220; iprobenfos; isofenphos; isoxathion; ivermectin; malathion; mecarbam; mesulfenphos;

metaldehyde; metolcarb; milbemectin; moxidectin; naled; NC 184; nithiazine; omethoate; oxamyl; oxydemethon M; oxydeprofos; permethrin; phenthroate; phorate; phosmet; phoxim; pirimiphos M; pirimiphos E; promecarb; propaphos; prothiofos; prothoate; pyrachlophos; pyradaphenthion; pyresmethrin; pyrethrum; tebufenozone; salithion; sebufos; sulfotep; sulprofos; tebufenpyrad; tebupirimphos; tefluthrin; temephos; terbam; tetrachlorvinphos; thiocloprid; thiafenox; thiamethoxam; thiodicarb; thiofanox; thionazin; thuringiensin; tralomethrin; triarathene; triazophos; triazuron; trichlorfon; triflumuron; trimethacarb; vamidothion; xylylcarb; YI 5301/5302; zetamethrin; DPX-MP062 — indoxacarb; methoxyfenozide; bifenazate; XMC (3,5-xylyl methylcarbamate); or the fungus pathogen *Metarhizium anisopliae*.

The compounds according to the invention can be used to control, i.e. to inhibit or destroy, pests of the mentioned type occurring on plants, especially on useful plants and ornamentals in agriculture, in horticulture and in forestry, or on parts of such plants, such as the fruits, blossoms, leaves, stems, tubers or roots, while in some cases plant parts that grow later are still protected against those pests.

Target crops include especially cereals, such as wheat, barley, rye, oats, rice, maize and sorghum; beet, such as sugar beet and fodder beet; fruit, e.g. pomes, stone fruit and soft fruit, such as apples, pears, plums, peaches, almonds, cherries and berries, e.g. strawberries, raspberries and blackberries; leguminous plants, such as beans, lentils, peas and soybeans; oil plants, such as rape, mustard, poppy, olives, sunflowers, coconut, castor oil, cocoa and groundnuts; cucurbitaceae, such as marrows, cucumbers and melons; fibre plants, such as cotton, flax, hemp and jute; citrus fruits, such as oranges, lemons, grapefruit and mandarins; vegetables, such as spinach, lettuce, asparagus, cabbages, carrots, onions, tomatoes, potatoes and paprika; lauraceae, such as avocado, cinnamon and camphor; and tobacco, nuts, coffee, aubergines, sugar cane, tea, pepper, vines, hops, bananas, natural rubber plants and ornamentals.

Further areas of use of the compounds according to the invention are the protection of stored goods and storerooms and the protection of raw materials, and also in the hygiene sector, especially the protection of domestic animals and productive livestock against pests of the mentioned type, more especially the protection of domestic animals, especially cats and dogs, from infestation by fleas, ticks and nematodes.

The invention therefore relates also to pesticidal compositions, such as emulsifiable concentrates, suspension concentrates, directly sprayable or dilutable solutions, spreadable

pastes, dilute emulsions, wettable powders, soluble powders, dispersible powders, wettable powders, dusts, granules and encapsulations of polymer substances, that comprise at least one of the compounds according to the invention, the choice of formulation being made in accordance with the intended objectives and the prevailing circumstances.

The active ingredient is used in those compositions in pure form; a solid active ingredient, for example, in a specific particle size, or preferably together with at least one of the adjuvants customary in formulation technology, such as extenders, e.g. solvents or solid carriers, or surface-active compounds (surfactants). In the area of parasite control in humans, domestic animals, productive livestock and pets it will be self-evident that only physiologically tolerable additives are used.

Solvents are, for example: non-hydrogenated or partly hydrogenated aromatic hydrocarbons, preferably fractions C₈ to C₁₂ of alkylbenzenes, such as xylene mixtures, alkylated naphthalenes or tetrahydronaphthalene, aliphatic or cycloaliphatic hydrocarbons, such as paraffins or cyclohexane, alcohols, such as ethanol, propanol or butanol, glycols and ethers and esters thereof, such as propylene glycol, dipropylene glycol ether, ethylene glycol or ethylene glycol monomethyl or -ethyl ether, ketones, such as cyclohexanone, isophorone or diacetone alcohol, strongly polar solvents, such as N-methylpyrrolid-2-one, dimethyl sulfoxide or N,N-dimethylformamide, water, non-epoxidized or epoxidized plant oils, such as non-epoxidized or epoxidized rapeseed, castor, coconut or soya oil, and silicone oils.

The solid carriers used, for example for dusts and dispersible powders, are as a rule natural rock powders, such as calcite, talc, kaolin, montmorillonite or attapulgite. Highly disperse silicic acids or highly disperse absorbent polymers can also be added to improve the physical properties. Granular adsorptive granule carriers are porous types, such as pumice, crushed brick, sepiolite or bentonite, and non-sorbent carrier materials are calcite or sand. A large number of granular materials of inorganic or organic nature can furthermore be used, in particular dolomite or comminuted plant residues.

Surface-active compounds are, depending on the nature of the active compound to be formulated, nonionic, cationic and/or anionic surfactants or surfactant mixtures with good emulsifying, dispersing and wetting properties. The surfactants listed below are to be regarded only as examples; many other surfactants which are customary in formulation technology and are suitable according to the invention are described in the relevant literature.

Nonionic surfactants are, in particular, polyglycol ether derivatives of aliphatic or cyclo-aliphatic alcohols, saturated or unsaturated fatty acids and alkylphenols, which can contain 3 to 30 glycol ether groups and 8 to 20 carbon atoms in the (aliphatic) hydrocarbon radical and 6 to 18 carbon atoms in the alkyl radical of the alkylphenols. Substances which are furthermore suitable are water-soluble polyethylene oxide adducts, containing 20 to 250 ethylene glycol ether and 10 to 100 propylene glycol ether groups, on propylene glycol, ethylene diaminopolypropylene glycol and alkyl polypropylene glycol having 1 to 10 carbon atoms in the alkyl chain. The compounds mentioned usually contain 1 to 5 ethylene glycol units per propylene glycol unit. Examples are nonylphenol-polyethoxyethanols, castor oil polyglycol ethers, polypropylene-polyethylene oxide adducts, tributylphenoxypropyl-ethoxyethanol, polyethylene glycol and octylphenoxypropyl-ethoxyethanol. Other substances are fatty acid esters of polyoxyethylene sorbitan, such as polyoxyethylene sorbitan trioleate.

The cationic surfactants are, in particular, quaternary ammonium salts which contain, as substituents, at least one alkyl radical having 8 to 22 C atoms and, as further substituents, lower, non-halogenated or halogenated alkyl, benzyl or lower hydroxyalkyl radicals. The salts are preferably in the form of halides, methyl-sulfates or ethyl-sulfates. Examples are stearyl-trimethyl-ammonium chloride and benzyl-di-(2-chloroethyl)-ethyl-ammonium bromide.

Suitable anionic surfactants can be both water-soluble soaps and water-soluble synthetic surface-active compounds. Suitable soaps are the alkali metal, alkaline earth metal and substituted or unsubstituted ammonium salts of higher fatty acids (C_{10} - C_{22}), such as the sodium or potassium salts of oleic or stearic acid, or of naturally occurring fatty acid mixtures, which can be obtained, for example, from coconut oil or tall oil; and furthermore also the fatty acid methyl-taurine salts. However, synthetic surfactants are more frequently used, in particular fatty sulfonates, fatty sulfates, sulfonated benzimidazole derivatives or alkylarylsulfonates. The fatty sulfonates and sulfates are as a rule in the form of alkali metal, alkaline earth metal or substituted or unsubstituted ammonium salts and in general have an alkyl radical of 8 to 22 C atoms, alkyl also including the alkyl moiety of acyl radicals; examples are the sodium or calcium salt of ligninsulfonic acid, of dodecylsulfuric acid ester or of a fatty alcohol sulfate mixture prepared from naturally occurring fatty acids. These also include the salts of sulfuric acid esters and sulfonic acids of fatty alcohol-ethylene oxide adducts. The sulfonated benzimidazole derivatives preferably contain 2 sulfonic acid groups and a fatty acid radical having about 8 to 22 C atoms. Alkylarylsulfonates are, for example, the sodium, calcium or triethanolammonium salts of dodecylbenzenesulfonic acid, of

dibutyl naphthalenesulfonic acid or of a naphthalenesulfonic acid-formaldehyde condensation product. Corresponding phosphates, such as salts of the phosphoric acid ester of a p-nonylphenol-(4-14)-ethylene oxide adduct or phospholipids, can further also be used.

The compositions as a rule comprise 0.1 to 99 %, in particular 0.1 to 95 %, of active compound and 1 to 99.9 %, in particular 5 to 99.9 %, of - at least - one solid or liquid auxiliary, it being possible as a rule for 0 to 25 %, in particular 0.1 to 20 %, of the composition to be surfactants (% is in each case per cent by weight). While concentrated compositions are more preferred as commercial goods, the end user as a rule uses dilute compositions which comprise considerably lower concentrations of active compound. Preferred compositions are composed, in particular, as follows (% = per cent by weight):

Emulsifiable concentrates:

active ingredient:	1 to 90%, preferably 5 to 20%
surfactant:	1 to 30%, preferably 10 to 20%
solvent:	5 to 98%, preferably 70 to 85%

Dusts:

active ingredient:	0.1 to 10%, preferably 0.1 to 1%
solid carrier:	99.9 to 90%, preferably 99.9 to 99%

Suspension concentrates:

active ingredient:	5 to 75%, preferably 10 to 50%
water:	94 to 24%, preferably 88 to 30%
surfactant:	1 to 40%, preferably 2 to 30%

Wettable powders:

active ingredient:	0.5 to 90%, preferably 1 to 80%
surfactant:	0.5 to 20%, preferably 1 to 15%
solid carrier:	5 to 99%, preferably 15 to 98%

Granules:

active ingredient:	0.5 to 30%, preferably 3 to 15%
solid carrier:	99.5 to 70%, preferably 97 to 85%

The compositions according to the invention may also comprise further solid or liquid adjuvants, such as stabilisers, e.g. vegetable oils or epoxidised vegetable oils (e.g. epoxi-

dised coconut oil, rapeseed oil or soybean oil), antifoams, e.g. silicone oil, preservatives, viscosity regulators, binders and/or tackifiers as well as fertilisers or other active ingredients for obtaining special effects, e.g. acaricides, bactericides, fungicides, nematicides, molluscicides or selective herbicides.

The crop protection products according to the invention are prepared in known manner, in the absence of adjuvants, e.g. by grinding, sieving and/or compressing a solid active ingredient or mixture of active ingredients, for example to a certain particle size, and in the presence of at least one adjuvant, for example by intimately mixing and/or grinding the active ingredient or mixture of active ingredients with the adjuvant(s). The invention relates likewise to those processes for the preparation of the compositions according to the invention and to the use of the compounds of formula (I) in the preparation of those compositions.

The invention relates also to the methods of application of the crop protection products, i.e. the methods of controlling pests of the mentioned type, such as spraying, atomising, dusting, coating, dressing, scattering or pouring, which are selected in accordance with the intended objectives and the prevailing circumstances, and to the use of the compositions for controlling pests of the mentioned type. Typical rates of concentration are from 0.1 to 1000 ppm, preferably from 0.1 to 500 ppm, of active ingredient. The rates of application per hectare are generally from 1 to 2000 g of active ingredient per hectare, especially from 10 to 1000 g/ha, preferably from 20 to 600 g/ha.

A preferred method of application in the area of crop protection is application to the foliage of the plants (foliar application), the frequency and the rate of application being dependent upon the risk of infestation by the pest in question. However, the active ingredient can also penetrate the plants through the roots (systemic action) when the locus of the plants is impregnated with a liquid formulation or when the active ingredient is incorporated in solid form into the locus of the plants, for example into the soil, e.g. in granular form (soil application). In the case of paddy rice crops, such granules may be applied in metered amounts to the flooded rice field.

The crop protection products according to the invention are also suitable for protecting plant propagation material, e.g. seed, such as fruits, tubers or grains, or plant cuttings, against animal pests. The propagation material can be treated with the composition before planting: seed, for example, can be dressed before being sown. The active ingredients according to the invention can also be applied to grains (coating), either by impregnating the seeds in a liquid formulation or by coating them with a solid formulation. The composition

can also be applied to the planting site when the propagation material is being planted, for example to the seed furrow during sowing. The invention relates also to such methods of treating plant propagation material and to the plant propagation material so treated.

The following Examples serve to illustrate the invention. They do not limit the invention. Temperatures are given in degrees Celsius; mixing ratios of solvents are given in parts by volume.

Preparation Examples

Example P1): Preparation of 1-(4-[3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy}-phenyl)-1H-pyrrole

P1.1: 2 g of methanesulfonic acid 3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propyl ester, 1.14 g of 4-iodophenol and 1.96 g of potassium carbonate are stirred for 24 hours at 50°C in 20 ml of dimethylformamide. The reaction mixture is poured onto water and extracted with ethyl acetate. The organic phase is washed with water and potassium carbonate solution and concentrated. Purification over silica gel yields 1,3-dichloro-5-(3,3-dichloro-allyloxy)-2-[3-(4-iodo-phenoxy)-propoxy]-benzene. ¹H-NMR (CDCl₃) 300 MHz: 2.28 (m,2H), 4.13 (t,2H), 4.22 (t,2H), 4.58 (d,2H), 6.10 (t,1H), 6.71 (d,2H), 6.81 (s,2H), 7.55 (d,2H)

P1.2: 200 mg of 1,3-dichloro-5-(3,3-dichloro-allyloxy)-2-[3-(4-iodo-phenoxy)-propoxy]-benzene, 29 mg of pyrrole, 6.9 mg of copper(I) iodide, 2.2 mg of ethylenediamine and 155 mg of potassium triphosphate are stirred for 24 hours at 110°C in 4 ml of dioxane. The reaction mixture is diluted with ethyl acetate and filtered. Concentration and purification over silica gel yield the title compound (compound 1.1).

Example P2): Preparation of 5-(4-[3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy}-phenyl)-2H-tetrazole and 5-(4-[3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy}-phenyl)-2-ethyl-2H-tetrazole

P2.1: 3 g of methanesulfonic acid 3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propyl ester, 0.93 g of 4-hydroxybenzonitrile and 2.9 g of potassium carbonate are stirred for 17 hours at 40°C in 30 ml of dimethylformamide. The reaction mixture is poured onto water and extracted with ethyl acetate. Concentration of the organic phase yields 4-(3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy)-benzonitrile. ¹H-NMR (CDCl₃) 300 MHz: 2.30 (m,2H), 4.14 (t,2H), 4.32 (t,2H), 4.58 (d,2H), 6.10 (t,1H), 6.83 (s,2H), 7.00 (d,2H), 7.60 (d,2H)

P2.2: 2 g of 4-{3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy}-benzonitrile, 1.2 g of ammonium chloride and 1.5 g of sodium azide are stirred for 48 hours at 100°C in 50 ml of dimethylformamide. The reaction mixture is poured onto cold dilute hydrochloric acid and extracted with ethyl acetate. Concentration of the organic phase and crystallisation from diethyl ether/hexane yield 5-(4-{3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy}-phenyl)-2H-tetrazole having a melting point of 159-161°C.

P2.3: 100 mg of 5-(4-{3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy}-phenyl)-2H-tetrazole, 48 mg of ethyl iodide and 141 mg of potassium carbonate are stirred for 4 hours at 50°C in 3 ml of dimethylformamide. The reaction mixture is poured onto water and extracted with ethyl acetate. Concentration of the organic phase and purification over silica gel yield 5-(4-{3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy}-phenyl)-2-ethyl-2H-tetrazole (compound 1.4).

Example P3: Preparation of 3-(4-{3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy}-phenyl)-5-methyl-[1,2,4]oxadiazole

P3.1: 1.1 g of 4-{3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy}-benzonitrile, 247 mg of hydroxylamine hydrochloride and 430 mg of triethylamine are stirred for 48 hours at 80°C in 25 ml of ethanol. The reaction mixture is poured onto water and extracted with ethyl acetate. Concentration of the organic phase and crystallisation from diethyl ether yield 4-{3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy}-N-hydroxy-benzamidine having a melting point of 144-146°C.

P3.2: 100 mg of 4-{3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy}-N-hydroxy-benzamidine are placed in 2 ml of toluene. 20 mg of acetyl chloride and 2 ml of pyridine are added, and heating is carried out for 4 hours at 120°C. The reaction mixture is concentrated, and the residue is taken up in ethyl acetate and washed with dilute hydrochloric acid and water. Concentration of the organic phase and purification over silica gel yield the title compound (compound 1.10).

Example P4: Preparation of 5-chloro-3-(4-{3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy}-phenyl)-isoxazole

P4.1: 307 mg of tert-butyl hypochloride in 5 ml of carbon tetrachloride are added dropwise over a period of 10 minutes, at 20-30°C, to 1.3 g of 4-{3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy}-N-hydroxy-benzimide in 15 ml of carbon tetrachloride. After 3 hours at room temperature, the reaction mixture is washed with water and concen-

trated. Crystallisation from hexane yields 4-{3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy}-N-hydroxy-benzimidoyl chloride.

P4.2: 150 mg of 4-{3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy}-N-hydroxy-benzimidoyl chloride are placed in 1 ml of 1,1-dichloroethane at 0°C, and a solution of 76 mg of triethylamine in 1 ml of 1,1-dichloroethane is added dropwise thereto over a period of one hour. After one hour at 0°C and 18 hours at 25°C, the reaction mixture is poured onto water and extracted with ethyl acetate. Concentration and purification over silica gel yield the title compound (compound 1.17).

Example P5): Preparation of 1-(4-{3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy}-phenyl)-5-isopropylsulfanyl-1H-tetrazole

P5.1: 388 mg of 4-(5-mercaptop-tetrazol-1-yl)-phenol, 430 mg of isopropyl iodide and 168 mg of sodium hydrogen carbonate are stirred for 5 hours at 80°C in 4 ml of acetonitrile. The reaction mixture is diluted with ethyl acetate, washed with sodium hydrogen carbonate solution and water, and concentrated. 4-(5-Isopropylsulfanyl-tetrazol-1-yl)-phenol is obtained in that manner. ¹H-NMR [(CD₃)₂SO] 300 MHz: 1.38 (d,6H), 3.78 (m,1H), 6.80 (d,2H), 7.25 (d,2H), 10.08 (s,1H).

P5.2: 400 mg of 4-(5-isopropylsulfanyl-tetrazol-1-yl)-phenol, 471 mg of bromopropanol, 473 mg of potassium carbonate and 10 mg of potassium iodide are stirred for 15 hours at reflux in 10 ml of acetone. The reaction mixture is filtered and concentrated. The residue is taken up in ethyl acetate, washed with water and concentrated. Purification over silica gel yields 3-[4-(5-isopropylsulfanyl-tetrazol-1-yl)-phenoxy]-propan-1-ol. ¹H-NMR (CDCl₃) 300 MHz: 1.50 (d,6H), 2.09 (m,2H), 3.90 (t,2H), 4.12 (m,1H), 4.19 (t,2H), 7.03 (d,2H), 7.43 (d,2H)

P5.3: 1.41 g of azodicarboxylic acid diisopropyl ester are added at 0°C to 1.83 g of triphenylphosphine in 30 ml of THF. After 30 minutes, a solution of 1.96 g of 3-[4-(5-isopropylsulfanyl-tetrazol-1-yl)-phenoxy]-propan-1-ol and 1.88 g of benzoic acid 3,5-dichloro-4-hydroxy-phenyl ester in 30 ml of THF is added dropwise. After one hour at 0°C and 24 hours at room temperature, the reaction mixture is concentrated. Purification over silica gel yields benzoic acid 3,5-dichloro-4-{3-[4-(5-isopropylsulfanyl-tetrazol-1-yl)-phenoxy]-propoxy}-phenyl ester. ¹H-NMR (CDCl₃) 300 MHz: 1.50 (d,2H), 2.33 (m,2H), 4.26 (t,2H), 4.38 (t,2H), 4.99 (m,1H), 7.09 (d,2H), 7.23 (s,2H), 7.42-7.70 (m,5H), 8.15 (d,2H)

P5.4: 3.5 ml of n-butylamine are added dropwise at 0-5°C to a solution of 2 g of benzoic acid 3,5-dichloro-4-{3-[4-(5-isopropylsulfanyl-tetrazol-1-yl)-phenoxy]-propoxy}-phenyl

ester. After 30 minutes at 0-5°C and one hour at room temperature, the reaction mixture is concentrated. Purification over silica gel yields 3,5-dichloro-4-[3-[4-(5-isopropylsulfanyl-tetrazol-1-yl)-phenoxy]-propoxy]-phenol. ¹H-NMR (CDCl₃) 300MHz: 1.50 (d,2H), 2.32 (m,2H), 4.08-4.20 (m,3H), 4.35 (t,2H), 5.62 (s,1H), 6.82 (s,2H), 7.09 (d,2H), 7.45 (d,2H)

P5.5: 900 mg of 3,5-dichloro-4-[3-[4-(5-isopropylsulfanyl-tetrazol-1-yl)-phenoxy]-propoxy]-phenol, 720 mg of 1,1,1,3-tetrachloropropane, 691 mg of potassium carbonate and 10 mg of potassium iodide are stirred for 48 hours at 60°C in 30 ml of acetone. The reaction mixture is filtered and the filtrate is concentrated. Purification over silica gel yields the title compound (compound 1.20).

Example P6: Preparation of 3-(4-[3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy]-phenyl)-4H-[1,2,4]oxadiazol-5-one

90 mg of chloroformic acid ethyl ester in 1 ml of acetone are added dropwise at 5°C to 400 mg of 4-[3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy]-N-hydroxy-benzamidine and 127 mg of potassium carbonate in 3 ml of acetone. After 3 hours at 0-5°C, the reaction mixture is poured onto water and extracted with dichloromethane. Concentration of the organic phase and purification over silica gel yield the title compound (compound 1.15).

Example P7: Preparation of 5-chloro-3-(4-[3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy]-phenyl)-[1,2,4]oxadiazole

90 mg of 3-(4-[3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy]-phenyl)-4H-[1,2,4]oxadiazol-5-one, 14 mg of pyridine and 0.2 ml of phosphorus oxychloride are stirred for 14 hours at 130°C. The reaction mixture is poured onto ice-water and extracted with ethyl acetate. Concentration of the organic phase and purification over silica gel yield the title compound (compound 1.16).

Example P8: Preparation of 3-(4-[3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy]-phenyl)-[1,2,4]oxadiazole

150 mg of 4-[3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy]-N-hydroxy-benzamidine and 15 mg of boron trifluoride ethyl etherate are stirred for 2 hours at 140°C in 2 ml of triethyl orthoformate. The reaction mixture is concentrated, and the residue is taken up in ethyl acetate and washed with water. Concentration of the organic phase and purification over silica gel yield the title compound (compound 1.14).

Example P9): Preparation of 2-(4-{3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy}-phenyl)-4-methyl-thiazole

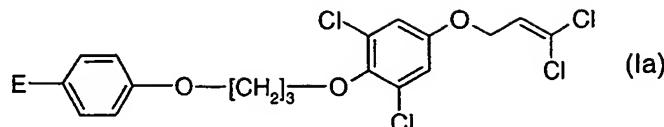
P9.1: Preparation of 4-(4-methyl-thiazol-2-yl)-phenol. 200 mg of 4-hydroxy-thiobenzamide and 144 mg of chloroacetone are stirred for 17 hours at room temperature and for 24 hours at 80°C in 2 ml of ethanol. The reaction mixture is concentrated completely. The title compound so obtained is reacted further in the form of the crude product.

P9.2: 96 mg of 4-(4-methyl-thiazol-2-yl)-phenol, 212 mg of methanesulfonic acid 3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propyl ester and 207 mg of potassium carbonate are stirred for 24 hours at 50°C in 3 ml of dimethylformamide. The reaction mixture is poured onto water and extracted with ethyl acetate. Concentration of the organic phase and purification over silica gel yield the title compound (compound 1.23).

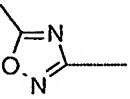
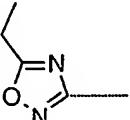
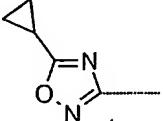
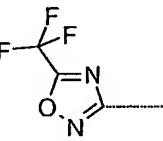
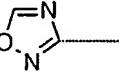
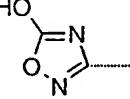
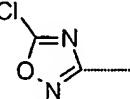
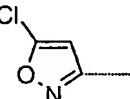
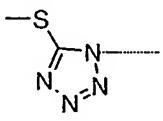
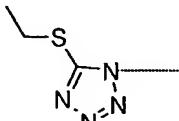
Example P10): Preparation of 1-(4-{3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propoxy}-phenyl)-1H-[1,2,4]triazole

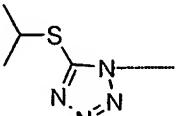
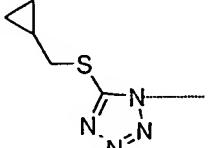
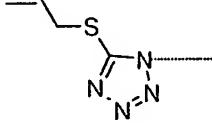
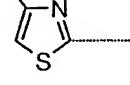
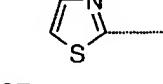
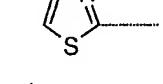
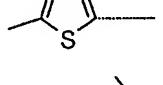
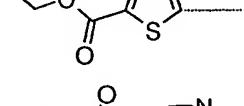
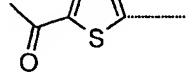
125 mg of 4-[1,2,4]triazol-1-yl-phenol, 212 mg of methanesulfonic acid 3-[2,6-dichloro-4-(3,3-dichloro-allyloxy)-phenoxy]-propyl ester and 207 mg of potassium carbonate are stirred for 24 hours at 50°C in 4 ml of dimethylformamide. The reaction mixture is poured onto water and extracted with ethyl acetate. Concentration of the organic phase and purification over silica gel yield the title compound (compound 1.9).

Example P11): In a manner analogous to that described above it is also possible to prepare the further compounds of the following Tables. The bond given with the symbol —— in the said tables indicates the connection of the substituent E with the remainder of the chemical structure.

Table 1: Compounds of formula

No.	E	¹ H-NMR (CDCl_3) 300MHz
1.1		2.30 (m,2H), 4.16 (t,2H), 4.28 (t,2H), 4.59 (d, 2H) 6.10 (t, 1H), 6.31 (s,2H), 6.82 (s, 2H), 6.93-7.01 (d+s, 4H), 7.30 (d, 2H)
1.2		Solvent ($\text{CD}_3)_2\text{SO}$: 2.24 (m,2H), 4.12 (t,2H), 4.31 (t,2H), 4.70 (d,2H), 6.50 (t,1H), 7.17 (s,2H), 7.20 (d,2H), 7.98 (d,2H)
1.3		2.32 (m,2H), 4.18 (t,2H), 4.33 (t,2H), 4.39 (s,3H), 4.58 (d,2H), 6.11 (t,1H), 6.82 (s,2H), 7.03 (d,2H), 8.07 (d,2H)
1.4		1.69 (t,3H), 2.32 (m,2H), 4.18 (t,2H), 4.32 (t,2H), 4.58 (d,2H), 4.68 (q,2H), 6.10 (t,1H), 6.82 (s,2H), 7.03 (d,2H), 8.08 (d,2H)
1.5		1.00 (t,3H), 2.10 (m,2H), 2.32 (m,2H), 4.18 (t,2H), 4.32 (t,2H), 4.53-4.67 (m,4H), 6.10 (t,1H), 6.82 (s,2H), 7.03 (d,2H), 8.08 (d,2H)
1.6		1.00 (d,6H), 2.23-2.53 (m,3H), 4.18 (t,2H), 4.32 (t,2H), 4.43(d2H), 4.58 (d,2H), 6.11 (t,1H), 6.82 (s,2H), 7.03 (d,2H), 8.08 (d,2H)
1.7		1.00 (d,6H), 1.53-1.71 (m1H), 1.96 (m,2H), 2.32 (m,2H), 4.18 (t,2H), 4.33 (t,2H), 4.58 (d,2H), 4.65 (t,2H), 6.11 (t,1H), 6.82 (s,2H), 7.03 (d,2H), 8.08 (d,2H)
1.8		0.89 (t,3H), 1.20-1.43 (m,6H), 1.93-2.12 (m,2H), 2.32 (m,2H), 4.18 (t,2H), 4.33 (t,2H), 4.52-4.69 (m,4H), 6.11 (t,1H), 6.82 (s,2H), 7.03 (d,2H), 8.08 (d,2H)
1.9		2.31 (m,2H), 4.17 (t,2H), 4.31 (t,2H), 4.59 (d,2H), 6.11 (t,1H), 6.83 (s,2H), 7.04 (d,2H), 7.58 (d,2H), 8.08 (s,1H), 8.45 (s,1H)

No.	E	¹ H-NMR (CDCl ₃) 300MHz
1.10		2.32 (m,2H), 2.62 (s,3H), 4.18 (t,2H), 4.32 (t,2H), 4.59 (d,2H), 6.10 (t,1H), 6.82 (s,2H), 7.01 (d,2H), 8.00 (d,2H)
1.11		1.42 (t,3H), 2.30 (m,2H), 2.97 (q,2H), 4.18 (t,2H), 4.32 (t,2H), 4.58 (d,2H), 6.10 (t,1H), 6.82 (s,2H), 7.01 (d,2H), 8.00 (d,2H)
1.12		1.13-1.36 (m,4H), 2.17-2.38 (m,4H), 4.18 (t,2H), 4.32 (t,2H), 4.58 (d,2H), 6.12 (t,1H), 6.82 (s,2H), 7.00 (d,2H), 7.98 (d,2H)
1.13		2.32 (m,2H), 4.19 (t,2H), 4.35 (t,2H), 4.58 (d,2H), 6.10 (t,1H), 6.83 (s,2H), 7.06 (d,2H), 8.06 (d,2H)
1.14		2.32 (m,2H), 4.18 (t,2H), 4.33 (t,2H), 4.58 (d,2H), 6.10 (t,1H), 6.83 (s,2H), 7.03 (d,2H), 8.07 (d,2H), 8.70 (s,1H)
1.15		Solvent (CD ₃) ₂ SO: 2.12 (m,2H), 4.00 (t,2H), 4.20 (t,2H), 4.59 (d,2H), 6.48 (t,1H), 7.02-7.10 (s+d,4H), 7.67 (d,2H), 12.30-12.95 (s,1H)
1.16		2.31 (m,2H), 4.18 (t,2H), 4.32 (t,2H), 4.58 (d,2H), 6.10 (t,1H), 6.82 (s,2H), 7.02 (d,2H), 7.98 (d,2H)
1.17		2.32 (m,2H), 4.18 (t,2H), 4.31 (t,2H), 4.59 (d,2H), 6.10 (t,1H), 6.42 (s,1H), 6.82 (s,2H), 7.00 (d,2H), 7.70 (d,2H)
1.18		2.32 (m,2H), 2.83 (s,3H), 4.18 (t,2H), 4.37 (t,2H), 4.60 (d,2H), 6.12 (t,1H), 6.83 (s,2H), 7.09 (d,2H), 7.47 (d,2H)
1.19		1.50 (t,3H), 2.32 (m,2H), 3.49 (q,2H), 4.18 (t,2H), 4.35 (t,2H), 4.59 (d,2H), 6.11 (t,1H), 6.83 (s,2H), 7.08 (d,2H), 7.47 (d,2H)

No.	E	¹ H-NMR (CDCl ₃) 300MHz
1.20		1.50 (d,6H), 2.32 (m,2H), 4.08-4.20 (m,4H), 4.33 (t,2H), 4.59 (d,2H), 6.11 (t,1H), 6.83 (s,2H), 7.08 (d,2H), 7.47 (d,2H)
1.21		0.38 (m,2H), 0.68 (m,2H), 1.28 (m,1H), 2.32 (m,2H), 3.45 (d,2H), 4.19 (t,2H), 4.33 (t,2H), 4.59 (d,2H), 6.11 (t,1H), 6.83 (s,2H), 7.08 (d,2H), 7.48 (d,2H)
1.22		2.33 (m,2H), 4.00 (t,2H), 4.35 (t,2H), 4.59 (d,2H), 5.21 (d,1H), 5.39 (d,1H), 5.92-6.07 (m,1H), 6.10 (t,1H), 6.83 (s,2H), 7.09 (d,2H), 7.47 (d,2H)
1.23		2.30 (m,2H), 2.50 (s,3H), 4.17 (t,2H), 4.30 (t,2H), 4.59 (d,2H), 6.10 (t,1H), 6.80 (s,1H), 6.82 (s,2H), 6.98 (d,2H), 7.88 (d,2H)
1.24		
1.25		
1.26		
1.27		
1.28		
1.29		

No.	E	¹ H-NMR (CDCl ₃) 300MHz
1.30		
1.31		
1.32		
1.33		
1.34		
1.35		
1.36		
1.37		
1.38		
1.39		

No.	E	$^1\text{H-NMR}$ (CDCl_3) 300MHz
1.40		
1.41		
1.42		
1.43		
1.44		
1.45		
1.46		
1.47		
1.48		
1.49		
1.50		
1.51		

No.	E	¹ H-NMR (CDCl ₃) 300MHz
1.52		
1.53		
1.54		
1.55		
1.56		
1.57		
1.58		2.20-2.40 (m, 8H), 4.15 (t,2H), 4.35 (t,2H), 4.59 (d,2H), 5.95 (s,1H), 6.10 (t,1H), 6.80 (s,2H), 6.95 (d,2H), 7.35 (d,2H)
1.59		2.2-2.4 (m,5H), 4.15 (t,2H), 4.35 (t,2H), 4.59 (d,2H), 6.10 (t,1H), 6.40 (s,1H), 6.80 (s,2H), 7.00 (d,2H), 7.35 (d,2H)
1.60		2.20-2.40 (m,5H), 4.15 (t,2H), 4.35 (t,2H), 4.59 (d,2H), 6.10 (t,1H), 6.55 (s,1H), 6.80 (s,2H), 6.95 (d,2H), 7.35 (d,2H)
1.61		

No.	E	¹ H-NMR (CDCl ₃) 300MHz
1.62		
1.63		
1.64		
1.65		
1.66		
1.67		
1.68		
1.69		
1.70		
1.71		
1.72		
1.73		

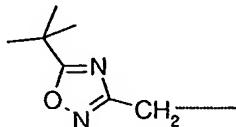
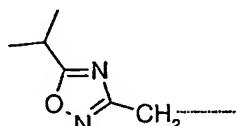
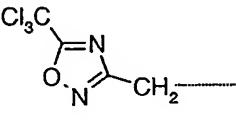
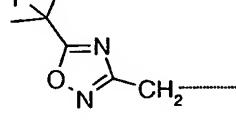
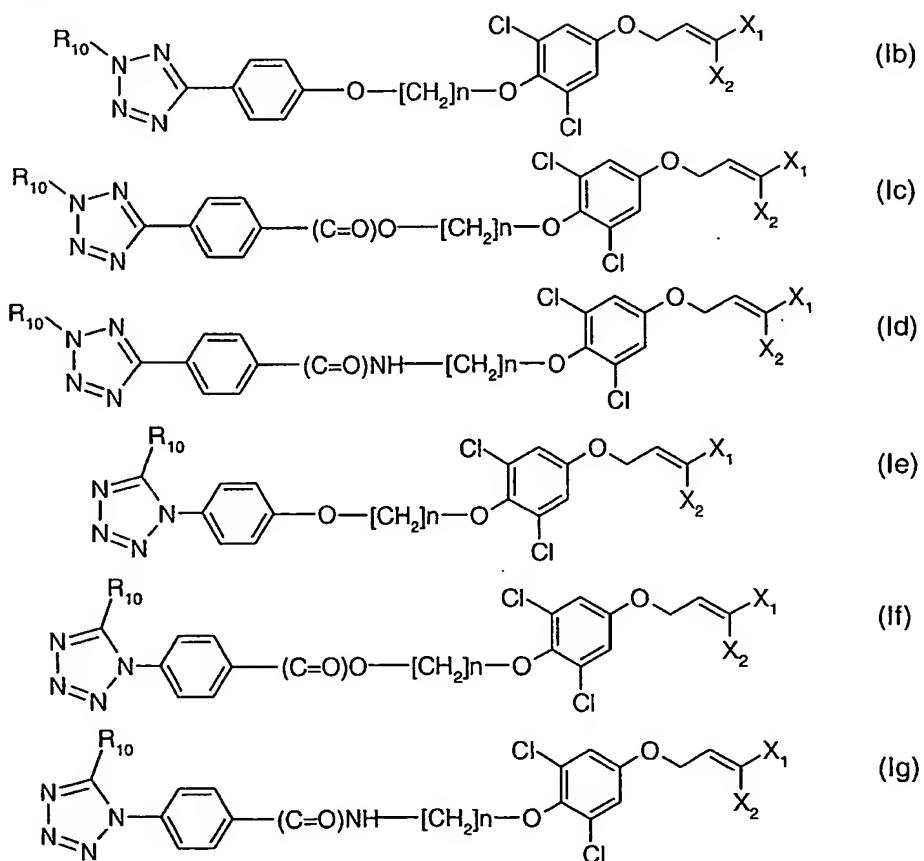
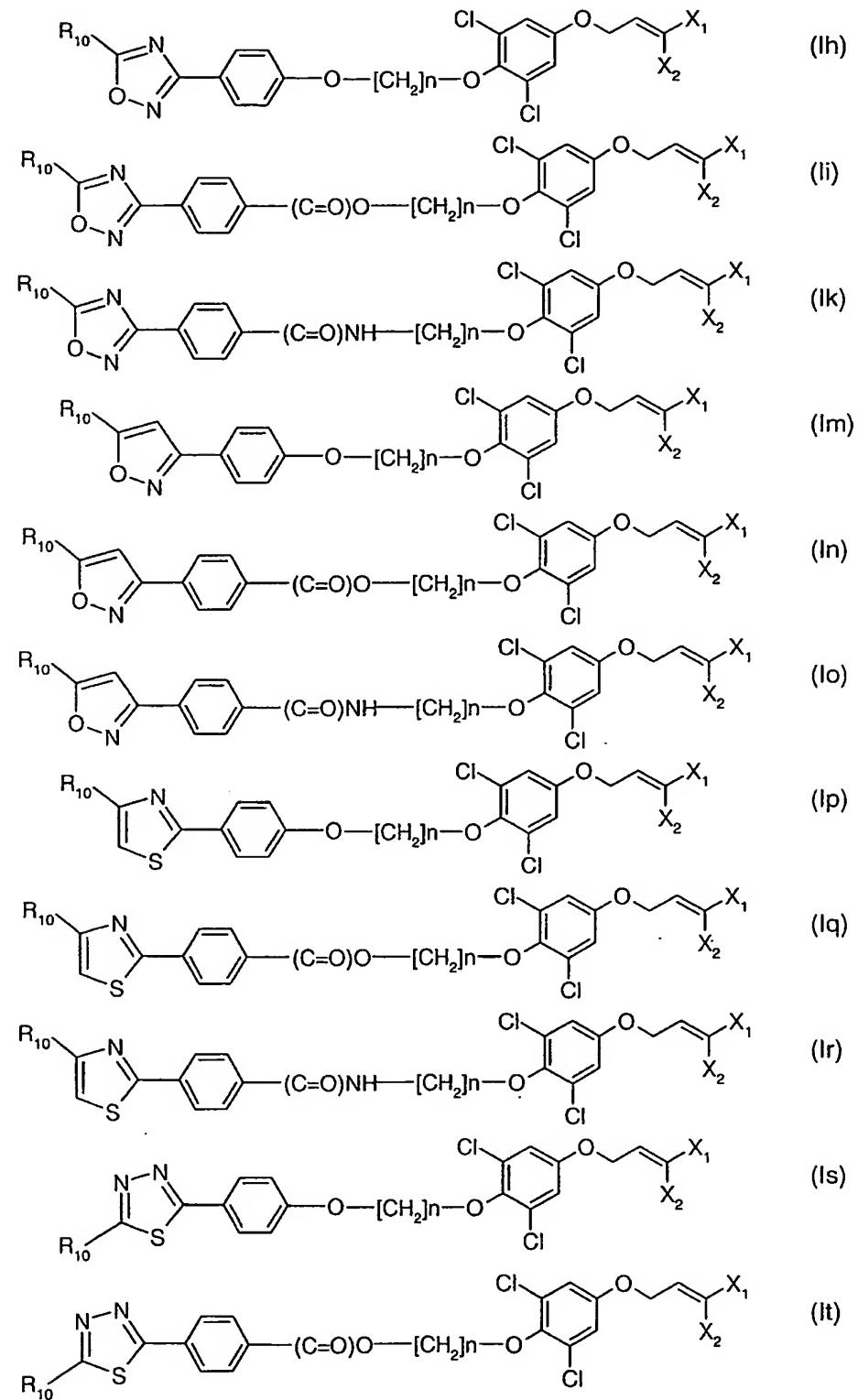
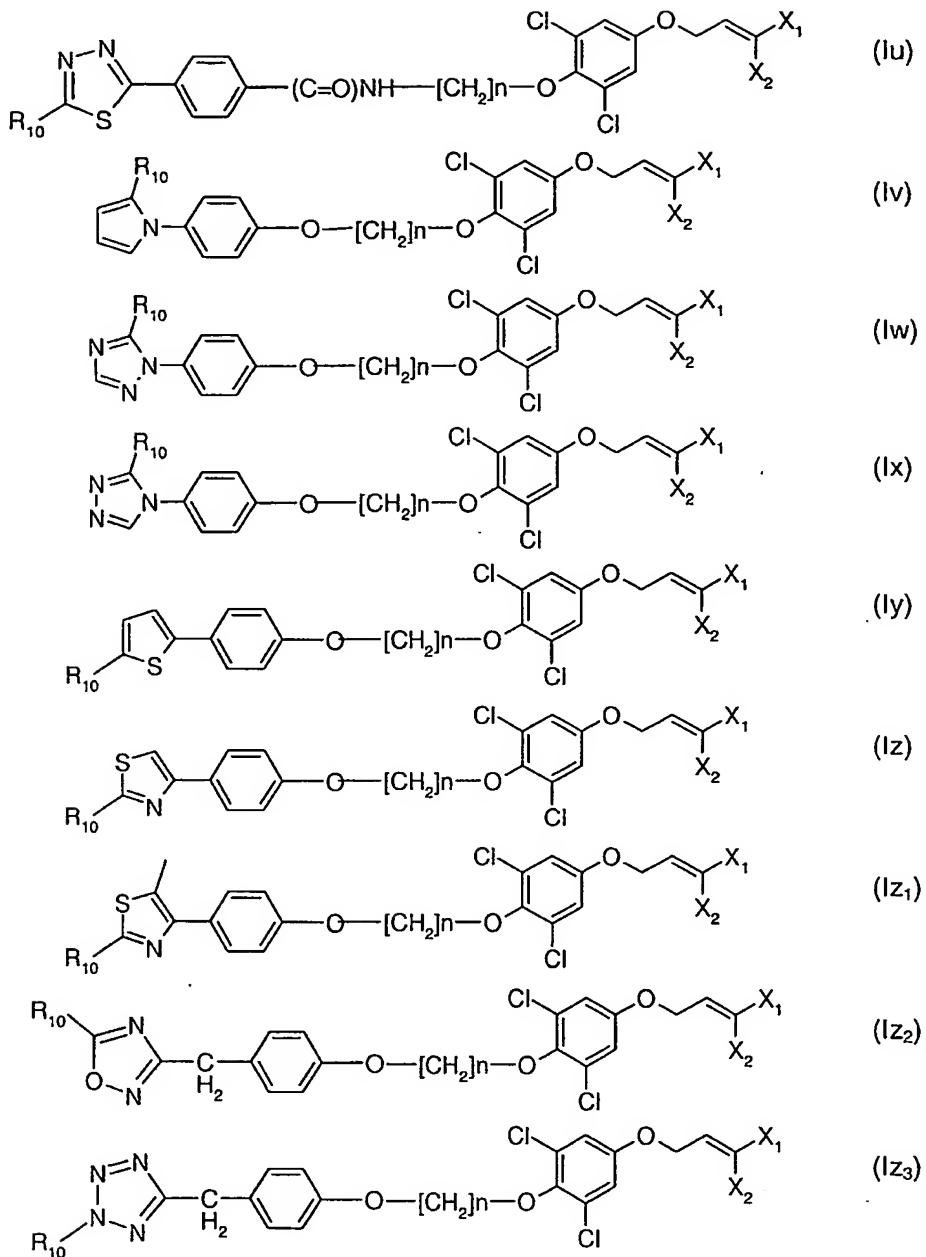
No.	E	¹ H-NMR (CDCl ₃) 300MHz
1.74		
1.75		
1.76		
1.77		

Table A: Compounds of formulae

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No.	X ₁	X ₂	R ₁₀
A.1	Cl	Cl	H
A.2	Cl	Cl	CN
A.3	Cl	Cl	Cl
A.4	Cl	Cl	Br
A.5	Cl	Cl	CH ₃

No.	X ₁	X ₂	R ₁₀
A.6	Cl	Cl	C ₂ H ₅
A.7	Cl	Cl	n-C ₃ H ₇
A.8	Cl	Cl	n-C ₄ H ₉
A.9	Cl	Cl	n-C ₅ H ₁₁
A.10	Cl	Cl	n-C ₆ H ₁₃
A.11	Cl	Cl	iso-C ₃ H ₇
A.12	Cl	Cl	iso-C ₄ H ₉
A.13	Cl	Cl	iso-C ₅ H ₁₁
A.14	Cl	Cl	tert-C ₄ H ₉
A.15	Cl	Cl	CH ₂ C(CH ₃) ₃
A.16	Cl	Cl	c-propyl
A.17	Cl	Cl	CH ₂ (c-propyl)
A.18	Cl	Cl	CF ₃
A.19	Cl	Cl	CH ₂ CF ₃
A.20	Cl	Cl	CH ₂ CHF ₂
A.21	Cl	Cl	CH ₂ CH ₂ F
A.22	Cl	Cl	CH ₂ CH=CH ₂
A.23	Cl	Cl	CH ₂ C≡CH
A.24	Cl	Cl	CH ₂ C≡CCH ₃
A.25	Cl	Cl	CH ₂ CH=CCl ₂
A.26	Cl	Cl	OH
A.27	Cl	Cl	OCH ₃
A.28	Cl	Cl	O-C ₂ H ₅
A.29	Cl	Cl	O-n-C ₃ H ₇
A.30	Cl	Cl	O-n-C ₄ H ₉
A.31	Cl	Cl	O-n-C ₅ H ₁₁
A.32	Cl	Cl	O-n-C ₆ H ₁₃
A.33	Cl	Cl	O-iso-C ₃ H ₇
A.34	Cl	Cl	O-iso-C ₄ H ₉
A.35	Cl	Cl	O-iso-C ₅ H ₁₁
A.36	Cl	Cl	O-tert-C ₄ H ₉
A.37	Cl	Cl	O-CH ₂ C(CH ₃) ₃
A.38	Cl	Cl	O-c-propyl
A.39	Cl	Cl	O-CH ₂ (c-propyl)
A.40	Cl	Cl	O-CF ₃
A.41	Cl	Cl	O-CH ₂ CF ₃
A.42	Cl	Cl	O-CH ₂ CHF ₂
A.43	Cl	Cl	O-CH ₂ CH ₂ F
A.44	Cl	Cl	O-CH ₂ CH=CH ₂
A.45	Cl	Cl	O-CH ₂ C≡CH
A.46	Cl	Cl	O-CH ₂ C≡CCH ₃
A.47	Cl	Cl	O-CH ₂ CH=CCl ₂

No.	X ₁	X ₂	R ₁₀
A.48	Cl	Cl	SH
A.49	Cl	Cl	SCH ₃
A.50	Cl	Cl	S-C ₂ H ₅
A.51	Cl	Cl	S-n-C ₃ H ₇
A.52	Cl	Cl	S-n-C ₄ H ₉
A.53	Cl	Cl	S-n-C ₅ H ₁₁
A.54	Cl	Cl	S-n-C ₆ H ₁₃
A.55	Cl	Cl	S-iso-C ₃ H ₇
A.56	Cl	Cl	S-iso-C ₄ H ₉
A.57	Cl	Cl	S-iso-C ₅ H ₁₁
A.58	Cl	Cl	S-tert-C ₄ H ₉
A.59	Cl	Cl	S-CH ₂ C(CH ₃) ₃
A.60	Cl	Cl	S-c-propyl
A.61	Cl	Cl	S-CH ₂ (c-propyl)
A.62	Cl	Cl	S-CF ₃
A.63	Cl	Cl	S-CH ₂ CF ₃
A.64	Cl	Cl	S-CH ₂ CHF ₂
A.65	Cl	Cl	S-CH ₂ CH ₂ F
A.66	Cl	Cl	S-CH ₂ CH=CH ₂
A.67	Cl	Cl	S-CH ₂ C≡CH
A.68	Cl	Cl	S-CH ₂ C=CCH ₃
A.69	Cl	Cl	S-CH ₂ CH=CCl ₂
A.70	Cl	Cl	CH ₂ OH
A.71	Cl	Cl	CH ₂ OCH ₃
A.72	Cl	Cl	CH ₂ O-C ₂ H ₅
A.73	Cl	Cl	CH ₂ O-n-C ₃ H ₇
A.74	Cl	Cl	CH ₂ O-n-C ₄ H ₉
A.75	Cl	Cl	CH ₂ O-n-C ₅ H ₁₁
A.76	Cl	Cl	CH ₂ O-n-C ₆ H ₁₃
A.77	Cl	Cl	CH ₂ O-iso-C ₃ H ₇
A.78	Cl	Cl	CH ₂ O-iso-C ₄ H ₉
A.79	Cl	Cl	CH ₂ O-iso-C ₅ H ₁₁
A.80	Cl	Cl	CH ₂ O-tert-C ₄ H ₉
A.81	Cl	Cl	CH ₂ O-CH ₂ C(CH ₃) ₃
A.82	Cl	Cl	CH ₂ O-c-propyl
A.83	Cl	Cl	CH ₂ O-CH ₂ (c-propyl)
A.84	Cl	Cl	CH ₂ O-CF ₃
A.85	Cl	Cl	CH ₂ O-CH ₂ CF ₃
A.86	Cl	Cl	CH ₂ O-CH ₂ CHF ₂
A.87	Cl	Cl	CH ₂ O-CH ₂ CH ₂ F
A.88	Cl	Cl	CH ₂ O-CH ₂ CH=CH ₂
A.89	Cl	Cl	CH ₂ O-CH ₂ C≡CH

No.	X ₁	X ₂	R ₁₀
A.90	Cl	Cl	CH ₂ O-CH ₂ C≡CCH ₃
A.91	Cl	Cl	CH ₂ O-CH ₂ CH=CCl ₂
A.92	Cl	Cl	C(=O)OH
A.93	Cl	Cl	C(=O)OCH ₃
A.94	Cl	Cl	C(=O)O-C ₂ H ₅
A.95	Cl	Cl	C(=O)O-n-C ₃ H ₇
A.96	Cl	Cl	C(=O)O-n-C ₄ H ₉
A.97	Cl	Cl	C(=O)O-n-C ₅ H ₁₁
A.98	Cl	Cl	C(=O)O-n-C ₆ H ₁₃
A.99	Cl	Cl	C(=O)O-iso-C ₃ H ₇
A.100	Cl	Cl	C(=O)O-iso-C ₄ H ₉
A.101	Cl	Cl	C(=O)O-iso-C ₅ H ₁₁
A.102	Cl	Cl	C(=O)O-tert-C ₄ H ₉
A.103	Cl	Cl	C(=O)O-CH ₂ C(CH ₃) ₃
A.104	Cl	Cl	C(=O)O-c-propyl
A.105	Cl	Cl	C(=O)O-CH ₂ (c-propyl)
A.106	Cl	Cl	C(=O)O-CF ₃
A.107	Cl	Cl	C(=O)O-CH ₂ CF ₃
A.108	Cl	Cl	C(=O)O-CH ₂ CHF ₂
A.109	Cl	Cl	C(=O)O-CH ₂ CH ₂ F
A.110	Cl	Cl	C(=O)O-CH ₂ CH=CH ₂
A.111	Cl	Cl	C(=O)O-CH ₂ C≡CH
A.112	Cl	Cl	C(=O)O-CH ₂ C≡CCH ₃
A.113	Cl	Cl	C(=O)O-CH ₂ CH=CCl ₂
A.114	Cl	Cl	C(=O)CH ₃
A.115	Cl	Cl	C(=O)-C ₂ H ₅
A.116	Cl	Cl	C(=O)-n-C ₃ H ₇
A.117	Cl	Cl	C(=O)-n-C ₄ H ₉
A.118	Cl	Cl	C(=O)-n-C ₅ H ₁₁
A.119	Cl	Cl	C(=O)-n-C ₆ H ₁₃
A.120	Cl	Cl	C(=O)-iso-C ₃ H ₇
A.121	Cl	Cl	C(=O)-iso-C ₄ H ₉
A.122	Cl	Cl	C(=O)-iso-C ₅ H ₁₁
A.123	Cl	Cl	C(=O)-tert-C ₄ H ₉
A.124	Cl	Cl	C(=O)-CH ₂ C(CH ₃) ₃
A.125	Cl	Cl	C(=O)-c-propyl
A.126	Cl	Cl	C(=O)-CH ₂ (c-propyl)
A.127	Cl	Cl	C(=O)-CF ₃
A.128	Cl	Cl	C(=O)-CH ₂ CF ₃
A.129	Cl	Cl	C(=O)-CH ₂ CHF ₂
A.130	Cl	Cl	C(=O)-CH ₂ CH ₂ F
A.131	Cl	Cl	C(=O)-CH ₂ CH=CH ₂

No.	X ₁	X ₂	R ₁₀
A.132	Cl	Cl	C(=O)-CH ₂ C≡CH
A.133	Cl	Cl	C(=O)-CH ₂ C≡CCH ₃
A.134	Cl	Cl	C(=O)-CH ₂ CH=CCl ₂
A.135	Br	Br	H
A.136	Br	Br	CN
A.137	Br	Br	Cl
A.138	Br	Br	Br
A.139	Br	Br	CH ₃
A.140	Br	Br	C ₂ H ₅
A.141	Br	Br	n-C ₃ H ₇
A.142	Br	Br	n-C ₄ H ₉
A.143	Br	Br	n-C ₅ H ₁₁
A.144	Br	Br	n-C ₆ H ₁₃
A.145	Br	Br	iso-C ₃ H ₇
A.146	Br	Br	iso-C ₄ H ₉
A.147	Br	Br	iso-C ₅ H ₁₁
A.148	Br	Br	tert-C ₄ H ₉
A.149	Br	Br	CH ₂ C(CH ₃) ₃
A.150	Br	Br	c-propyl
A.151	Br	Br	CH ₂ (c-propyl)
A.152	Br	Br	CF ₃
A.153	Br	Br	CH ₂ CF ₃
A.154	Br	Br	CH ₂ CHF ₂
A.155	Br	Br	CH ₂ CH ₂ F
A.156	Br	Br	CH ₂ CH=CH ₂
A.157	Br	Br	CH ₂ C≡CH
A.158	Br	Br	CH ₂ C≡CCH ₃
A.159	Br	Br	CH ₂ CH=CCl ₂
A.160	Br	Br	OH
A.161	Br	Br	OCH ₃
A.162	Br	Br	O-C ₂ H ₅
A.163	Br	Br	O-n-C ₃ H ₇
A.164	Br	Br	O-n-C ₄ H ₉
A.165	Br	Br	O-n-C ₅ H ₁₁
A.166	Br	Br	O-n-C ₆ H ₁₃
A.167	Br	Br	O-iso-C ₃ H ₇
A.168	Br	Br	O-iso-C ₄ H ₉
A.169	Br	Br	O-iso-C ₅ H ₁₁
A.170	Br	Br	O-tert-C ₄ H ₉
A.171	Br	Br	O-CH ₂ C(CH ₃) ₃
A.172	Br	Br	O-c-propyl
A.173	Br	Br	O-CH ₂ (c-propyl)

No.	X ₁	X ₂	R ₁₀
A.174	Br	Br	O-CF ₃
A.175	Br	Br	O-CH ₂ CF ₃
A.176	Br	Br	O-CH ₂ CHF ₂
A.177	Br	Br	O-CH ₂ CH ₂ F
A.178	Br	Br	O-CH ₂ CH=CH ₂
A.179	Br	Br	O-CH ₂ C≡CH
A.180	Br	Br	O-CH ₂ C≡CCH ₃
A.181	Br	Br	O-CH ₂ CH=CCl ₂
A.182	Br	Br	SH
A.183	Br	Br	SCH ₃
A.184	Br	Br	S-C ₂ H ₅
A.185	Br	Br	S-n-C ₃ H ₇
A.186	Br	Br	S-n-C ₄ H ₉
A.187	Br	Br	S-n-C ₅ H ₁₁
A.188	Br	Br	S-n-C ₆ H ₁₃
A.189	Br	Br	S-iso-C ₃ H ₇
A.190	Br	Br	S-iso-C ₄ H ₉
A.191	Br	Br	S-iso-C ₅ H ₁₁
A.192	Br	Br	S-tert-C ₄ H ₉
A.193	Br	Br	S-CH ₂ C(CH ₃) ₃
A.194	Br	Br	S-c-propyl
A.195	Br	Br	S-CH ₂ (c-propyl)
A.196	Br	Br	S-CF ₃
A.197	Br	Br	S-CH ₂ CF ₃
A.198	Br	Br	S-CH ₂ CHF ₂
A.199	Br	Br	S-CH ₂ CH ₂ F
A.200	Br	Br	S-CH ₂ CH=CH ₂
A.201	Br	Br	S-CH ₂ C≡CH
A.202	Br	Br	S-CH ₂ C≡CCH ₃
A.203	Br	Br	S-CH ₂ CH=CCl ₂
A.204	Br	Br	CH ₂ OH
A.205	Br	Br	CH ₂ OCH ₃
A.206	Br	Br	CH ₂ O-C ₂ H ₅
A.207	Br	Br	CH ₂ O-n-C ₃ H ₇
A.208	Br	Br	CH ₂ O-n-C ₄ H ₉
A.209	Br	Br	CH ₂ O-n-C ₅ H ₁₁
A.210	Br	Br	CH ₂ O-n-C ₆ H ₁₃
A.211	Br	Br	CH ₂ O-iso-C ₃ H ₇
A.212	Br	Br	CH ₂ O-iso-C ₄ H ₉
A.213	Br	Br	CH ₂ O-iso-C ₅ H ₁₁
A.214	Br	Br	CH ₂ O-tert-C ₄ H ₉
A.215	Br	Br	CH ₂ O-CH ₂ C(CH ₃) ₃

No.	X ₁	X ₂	R ₁₀
A.216	Br	Br	CH ₂ O-c-propyl
A.217	Br	Br	CH ₂ O-CH ₂ (c-propyl)
A.218	Br	Br	CH ₂ O-CF ₃
A.219	Br	Br	CH ₂ O-CH ₂ CF ₃
A.220	Br	Br	CH ₂ O-CH ₂ CHF ₂
A.221	Br	Br	CH ₂ O-CH ₂ CH ₂ F
A.222	Br	Br	CH ₂ O-CH ₂ CH=CH ₂
A.223	Br	Br	CH ₂ O-CH ₂ C≡CH
A.224	Br	Br	CH ₂ O-CH ₂ C≡CCH ₃
A.225	Br	Br	CH ₂ O-CH ₂ CH=CCl ₂
A.226	Br	Br	C(=O)OH
A.227	Br	Br	C(=O)OCH ₃
A.228	Br	Br	C(=O)O-C ₂ H ₅
A.229	Br	Br	C(=O)O-n-C ₃ H ₇
A.230	Br	Br	C(=O)O-n-C ₄ H ₉
A.231	Br	Br	C(=O)O-n-C ₅ H ₁₁
A.232	Br	Br	C(=O)O-n-C ₆ H ₁₃
A.233	Br	Br	C(=O)O-iso-C ₃ H ₇
A.234	Br	Br	C(=O)O-iso-C ₄ H ₉
A.235	Br	Br	C(=O)O-iso-C ₅ H ₁₁
A.236	Br	Br	C(=O)O-tert-C ₄ H ₉
A.237	Br	Br	C(=O)O-CH ₂ C(CH ₃) ₃
A.238	Br	Br	C(=O)O-c-propyl
A.239	Br	Br	C(=O)O-CH ₂ (c-propyl)
A.240	Br	Br	C(=O)O-CF ₃
A.241	Br	Br	C(=O)O-CH ₂ CF ₃
A.242	Br	Br	C(=O)O-CH ₂ CHF ₂
A.243	Br	Br	C(=O)O-CH ₂ CH ₂ F
A.244	Br	Br	C(=O)O-CH ₂ CH=CH ₂
A.245	Br	Br	C(=O)O-CH ₂ C≡CH
A.246	Br	Br	C(=O)O-CH ₂ C≡CCH ₃
A.247	Br	Br	C(=O)O-CH ₂ CH=CCl ₂
A.248	Br	Br	C(=O)CH ₃
A.249	Br	Br	C(=O)-C ₂ H ₅
A.250	Br	Br	C(=O)-n-C ₃ H ₇
A.251	Br	Br	C(=O)-n-C ₄ H ₉
A.252	Br	Br	C(=O)-n-C ₅ H ₁₁
A.253	Br	Br	C(=O)-n-C ₆ H ₁₃
A.254	Br	Br	C(=O)-iso-C ₃ H ₇
A.255	Br	Br	C(=O)-iso-C ₄ H ₉
A.256	Br	Br	C(=O)-iso-C ₅ H ₁₁
A.257	Br	Br	C(=O)-tert-C ₄ H ₉

No.	X ₁	X ₂	R ₁₀
A.258	Br	Br	C(=O)-CH ₂ C(CH ₃) ₃
A.259	Br	Br	C(=O)-c-propyl
A.260	Br	Br	C(=O)-CH ₂ (c-propyl)
A.261	Br	Br	C(=O)-CF ₃
A.262	Br	Br	C(=O)-CH ₂ CF ₃
A.263	Br	Br	C(=O)-CH ₂ CHF ₂
A.264	Br	Br	C(=O)-CH ₂ CH ₂ F
A.265	Br	Br	C(=O)-CH ₂ CH=CH ₂
A.266	Br	Br	C(=O)-CH ₂ C≡CH
A.267	Br	Br	C(=O)-CH ₂ C≡CCH ₃
A.268	Br	Br	C(=O)-CH ₂ CH=CCl ₂

Table 2: A compound of the general formula (Ib) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 3: A compound of the general formula (Ib) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 4: A compound of the general formula (Ib) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 5: A compound of the general formula (Ib) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 6: A compound of the general formula (Ic) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 7: A compound of the general formula (Ic) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 8: A compound of the general formula (Ic) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 9: A compound of the general formula (Ic) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 10: A compound of the general formula (Id) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 11: A compound of the general formula (Id) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 12: A compound of the general formula (Id) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 13: A compound of the general formula (Id) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 14: A compound of the general formula (Ie) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 15: A compound of the general formula (Ie) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 16: A compound of the general formula (Ie) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 17: A compound of the general formula (Ie) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 18: A compound of the general formula (If) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 19: A compound of the general formula (If) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 20: A compound of the general formula (If) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 21: A compound of the general formula (If) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 22: A compound of the general formula (Ig) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 23: A compound of the general formula (Ig) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 24: A compound of the general formula (Ig) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 25: A compound of the general formula (Ig) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 26: A compound of the general formula (Ih) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 27: A compound of the general formula (Ih) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 28: A compound of the general formula (Ih) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 29: A compound of the general formula (Ih) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 30: A compound of the general formula (II) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 31: A compound of the general formula (li) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 32: A compound of the general formula (li) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 33: A compound of the general formula (li) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 34: A compound of the general formula (Ik) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 35: A compound of the general formula (Ik) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 36: A compound of the general formula (Ik) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 37: A compound of the general formula (Ik) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 38: A compound of the general formula (Im) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 39: A compound of the general formula (Im) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 40: A compound of the general formula (Im) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 41: A compound of the general formula (Im) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 42: A compound of the general formula (In) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 43: A compound of the general formula (In) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 44: A compound of the general formula (In) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 45: A compound of the general formula (In) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 46: A compound of the general formula (Io) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 47: A compound of the general formula (Io) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 48: A compound of the general formula (Io) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 49: A compound of the general formula (Io) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 50: A compound of the general formula (Ip) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 51: A compound of the general formula (Ip) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 52: A compound of the general formula (Ip) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 53: A compound of the general formula (Ip) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 54: A compound of the general formula (Iq) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 55: A compound of the general formula (Iq) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 56: A compound of the general formula (Iq) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 57: A compound of the general formula (Iq) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 58: A compound of the general formula (Ir) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 59: A compound of the general formula (Ir) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 60: A compound of the general formula (Ir) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 61: A compound of the general formula (Ir) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 62: A compound of the general formula (Is) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 63: A compound of the general formula (Is) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 64: A compound of the general formula (Is) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 65: A compound of the general formula (Is) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 66: A compound of the general formula (It) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 67: A compound of the general formula (It) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 68: A compound of the general formula (It) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 69: A compound of the general formula (It) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 70: A compound of the general formula (Iu) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 71: A compound of the general formula (Iu) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 72: A compound of the general formula (Iu) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 73: A compound of the general formula (Iu) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 74: A compound of the general formula (Iv) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 75: A compound of the general formula (Iv) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 76: A compound of the general formula (Iv) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 77: A compound of the general formula (Iv) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 78: A compound of the general formula (Iw) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 79: A compound of the general formula (Iw) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 80: A compound of the general formula (Iw) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 81: A compound of the general formula (Iw) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 82: A compound of the general formula (Ix) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 83: A compound of the general formula (Ix) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 84: A compound of the general formula (Ix) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 85: A compound of the general formula (Ix) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 86: A compound of the general formula (Iy) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 87: A compound of the general formula (Iy) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 88: A compound of the general formula (Iy) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 89: A compound of the general formula (Iy) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 90: A compound of the general formula (Iz) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 91: A compound of the general formula (Iz) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 92: A compound of the general formula (Iz) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 93: A compound of the general formula (Iz) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 94: A compound of the general formula (Iz₁) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 95: A compound of the general formula (Iz₁) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 96: A compound of the general formula (Iz₁) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 97: A compound of the general formula (Iz₁) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 98: A compound of the general formula (Iz₂) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 99: A compound of the general formula (Iz₂) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 100: A compound of the general formula (Iz₂) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 101: A compound of the general formula (Iz₂) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 102: A compound of the general formula (Iz₃) wherein n is 2 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 103: A compound of the general formula (Iz₃) wherein n is 3 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 104: A compound of the general formula (Iz₃) wherein n is 4 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Table 105: A compound of the general formula (Iz₃) wherein n is 5 and the combination of the substituents R₁₀, X₁ and X₂ for each compound corresponds to a line A.1 to A.268 of Table A.

Formulation Examples (% = percent by weight)

<u>Example F1: Emulsifiable concentrates</u>	a)	b)	c)
active ingredient	25 %	40 %	50 %
calcium dodecylbenzenesulfonate	5 %	8 %	6 %
castor oil polyethylene glycol ether (36 mol EO)	5 %	-	-
tributylphenol polyethylene glycol ether (30 mol EO)	-	12 %	4 %
cyclohexanone	-	15 %	20 %
xylene mixture	65 %	25 %	20 %

Mixing finely ground active ingredient and additives gives an emulsifiable concentrate which yields emulsions of the desired concentration on dilution with water.

<u>Example F2: Solutions</u>	a)	b)	c)	d)
active ingredient	80 %	10 %	5 %	95 %
ethylene glycol monomethyl ether	20 %	-	-	-
polyethylene glycol (MW 400)	-	70 %	-	-
N-methylpyrrolid-2-one	-	20 %	-	-
epoxidised coconut oil	-	-	1 %	5 %
benzine (boiling range: 160-190°)	-	-	94 %	-

Mixing finely ground active ingredient and additives gives a solution suitable for use in the form of microdrops.

<u>Example F3: Granules</u>	a)	b)	c)	d)
active ingredient	5 %	10 %	8 %	21 %
kaolin	94 %	-	79 %	54 %
highly dispersed silicic acid	1 %	-	13 %	7 %
attapulgite	-	90 %	-	18 %

The active ingredient is dissolved in dichloromethane, the solution is sprayed onto the carrier mixture and the solvent is evaporated off *in vacuo*.

Biological ExamplesExample B1: Action against Heliothis virescens caterpillars

Young soybean plants are sprayed with an aqueous emulsion spray mixture comprising 400 ppm of test compound. After the spray-coating has dried, the soybean plants are populated with 10 caterpillars of *Heliothis virescens* in the first stage and placed in a plastics container. Evaluation is made 6 days later. The percentage reduction in population and the

percentage reduction in feeding damage (% activity) are determined by comparing the number of dead caterpillars and the feeding damage on the treated plants with that on the untreated plants.

The compounds of the Tables exhibit good activity against *Heliothis virescens* in this test. In particular, the compounds 1.3, 1.4, 1.5, 1.10 to 1.13, 1.16, 1.17, 1.19 and 1.21 are more than 80 % effective.

Example B2: Action against *Plutella xylostella* caterpillars

Young cabbage plants are sprayed with an aqueous emulsion spray mixture comprising 400 ppm of test compound. After the spray-coating has dried, the cabbage plants are populated with 10 caterpillars of *Plutella xylostella* in the third stage and placed in a plastics container. Evaluation is made 3 days later. The percentage reduction in population and the percentage reduction in feeding damage (% activity) are determined by comparing the number of dead caterpillars and the feeding damage on the treated plants with that on the untreated plants.

The compounds of the Tables exhibit good activity against *Plutella xylostella*. In particular, the compounds 1.3, 1.4, 1.5, 1.10 to 1.13, 1.16, 1.17, 1.19 and 1.21 are more than 80 % effective.

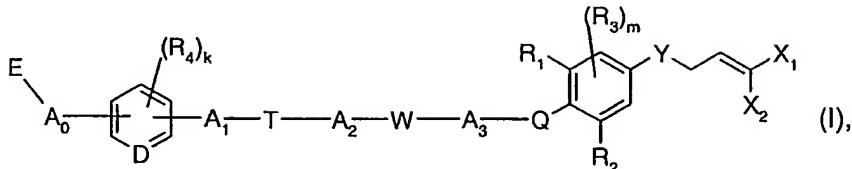
Example B3: Action against *Spodoptera littoralis*

Young soybean plants are sprayed with an aqueous emulsion spray mixture comprising 400 ppm of test compound and, after the spray-coating has dried, the plants are populated with 10 caterpillars of *Spodoptera littoralis* in the first stage and then placed in a plastics container. 3 days later, the percentage reduction in population and the percentage reduction in feeding damage (% activity) are determined by comparing the number of dead caterpillars and the feeding damage on the treated plants with that on untreated plants.

The compounds of the Tables exhibit good activity in this test. In particular, the compounds 1.3, 1.4, 1.5, 1.10 to 1.13, 1.16, 1.17, 1.19 and 1.21 are more than 80 % effective.

What is claimed is:

1. A compound of formula



wherein

A_0 , A_1 and A_2 are each independently of the other a bond or a C_1 - C_6 alkylene bridge which is unsubstituted or substituted by from one to six identical or different substituents selected from C_3 - C_8 cycloalkyl, C_3 - C_8 cycloalkyl- C_1 - C_6 alkyl and C_1 - C_3 haloalkyl;

A_3 is a C_1 - C_6 alkylene bridge which is unsubstituted or substituted by from one to six identical or different substituents selected from C_3 - C_8 cycloalkyl, C_3 - C_8 cycloalkyl- C_1 - C_6 alkyl and C_1 - C_3 haloalkyl;

D is CH or N;

X_1 and X_2 are each independently of the other fluorine, chlorine or bromine;

R_1 , R_2 and R_3 are each independently of the others H, halogen, OH, SH, CN, nitro, C_1 - C_6 alkyl, C_1 - C_6 haloalkyl, C_1 - C_6 alkylcarbonyl, C_2 - C_6 alkenyl, C_2 - C_6 haloalkenyl, C_2 - C_6 alkynyl, C_1 - C_6 alkoxy, C_1 - C_6 haloalkoxy, C_2 - C_6 alkenyloxy, C_2 - C_6 haloalkenyloxy, C_2 - C_6 alkynyloxy, $-S(=O)-C_1-C_6$ alkyl, $-S(O)_2-C_1-C_6$ alkyl, C_1-C_6 alkoxycarbonyl or C_3-C_6 haloalkynyloxy; the substituents R_3 being independent of one another when m is 2;

R_4 is H, halogen, OH, SH, CN, nitro, C_1 - C_6 alkyl, C_1 - C_6 haloalkyl, C_1 - C_6 alkylcarbonyl, C_2 - C_6 alkenyl, C_2 - C_6 haloalkenyl, C_2 - C_6 alkynyl, C_1 - C_6 alkoxy, C_1 - C_6 haloalkoxy, C_2 - C_6 alkenyl- oxy , C_2 - C_6 haloalkenyl- oxy , C_2 - C_6 alkenyloxy, $-S(=O)-C_1-C_6$ alkyl, $-S(O)_2-C_1-C_6$ alkyl or C_1-C_6 alkoxycarbonyl; the substituents R_4 being independent of one another when k is greater than 1; or $N(R_5)_2$ wherein the two substituents R_5 are independent of one another;

R_5 is H, CN, OH, C_1 - C_6 alkyl, C_3 - C_8 cycloalkyl, C_3 - C_8 cycloalkyl- C_1 - C_6 alkyl, C_1 - C_6 haloalkyl, C_1 - C_6 alkoxy, C_1 - C_6 haloalkoxy, C_2 - C_6 alkenyloxy, C_2 - C_6 haloalkenyloxy, C_2 - C_6 alkynyloxy, $-C(=O)R_8$, $-C(=S)R_8$, phenyl, benzyl; or phenyl or benzyl each of which is substituted in the aromatic ring by from one to five identical or different substituents selected from the group consisting of halogen, C_1 - C_6 alkyl, halo- C_1 - C_6 alkyl, C_1 - C_6 alkoxy, halo- C_1 - C_6 alkoxy, hydroxy, cyano and nitro;

or the two substituents R₅ together form a four- to eight-membered, straight-chain or branched alkylene bridge wherein a CH₂ group may have been replaced by O, S or NR₉, and the alkylene bridge is unsubstituted or substituted by from one to four identical or different substituents selected from C₃-C₈cycloalkyl, C₃-C₈cycloalkyl-C₁-C₆alkyl and C₁-C₃haloalkyl;

W is O, NR₆, S, SO, SO₂, -C(=O)-O-, -O-C(=O)-, -C(=O)-NR₇- or -NR₇-C(=O)-;

T is a bond, O, NH, NR₆, S, SO, SO₂, -C(=O)-O-, -O-C(=O)-, -C(=O)-NR₇- or -NR₇-C(=O)-;

Q is O, NR₆, S, SO or SO₂;

Y is O, NR₆, S, SO or SO₂;

R₆ and R₇ are independently of each other H, C₁-C₆alkyl, C₁-C₃haloalkyl, C₁-C₆alkyl-carbonyl, C₁-C₃haloalkylcarbonyl, C₁-C₆alkoxyalkyl, C₃-C₈cycloalkyl or benzyl;

R₈ is C₁-C₆alkyl, C₁-C₆haloalkyl, C₂-C₆alkenyl, C₂-C₆haloalkenyl, C₂-C₆alkynyl, C₁-C₆alkoxy, C₁-C₆haloalkoxy, C₂-C₆alkenyloxy, C₂-C₆haloalkenyloxy, C₂-C₆alkynyloxy, C₃-C₈cycloalkyl, phenyl, benzyl; or phenyl or benzyl each of which is unsubstituted or substituted by from one to three identical or different substituents selected from halogen, CN, nitro, C₁-C₆alkyl, C₁-C₆haloalkyl, C₁-C₆alkylcarbonyl, C₂-C₆alkenyl, C₂-C₆haloalkenyl, C₂-C₆alkynyl, C₁-C₆alkoxy, C₁-C₆haloalkoxy, C₁-C₆alkoxycarbonyl, C₁-C₃haloalkoxycarbonyl and C₂-C₆haloalkenyloxy;

R₉ is H, C₁-C₆alkyl, C₁-C₃haloalkyl, C₁-C₆-alkylcarbonyl, C₁-C₆haloalkylcarbonyl, C₁-C₆alkoxyalkyl, C₃-C₈cycloalkyl or benzyl;

k is 1, 2 or 3 when D is nitrogen; or is 1, 2, 3 or 4 when D is CH;

m is 1 or 2;

E is heteroaryl which is unsubstituted or substituted - depending upon the substitutions possible on the ring - by from one to four identical or different substituents selected from R₁₀;

R₁₀ is halogen, CN, NO₂, OH, SH, C₁-C₆alkyl, C₁-C₆haloalkyl, C₁-C₆hydroxyalkyl, C₃-C₈cycloalkyl, C₃-C₈cycloalkyl-C₁-C₆alkyl, C₂-C₆alkenyl, C₂-C₆haloalkenyl, C₃-C₆alkynyl, C₃-C₆haloalkynyl, C₁-C₆alkoxy, C₁-C₆alkoxy-C₁-C₆alkyl, C₁-C₆haloalkoxy, C₁-C₆haloalkoxy-C₁-C₆alkyl, C₂-C₆alkenyloxy, C₂-C₆haloalkenyloxy, C₂-C₆alkenyloxy-C₁-C₆alkyl, C₂-C₆haloalkenyloxy-C₁-C₆alkyl, C₃-C₆alkynyoxy, C₃-C₆haloalkynyoxy, C₃-C₆alkynyoxy-C₁-C₆alkyl, C₃-C₈cycloalkoxy, C₃-C₈cycloalkyl-C₁-C₆alkoxy, C₃-C₈cycloalkoxy-C₁-C₆alkyl, C₃-C₈cycloalkoxy-C₁-C₆alkoxy, C₃-C₈cycloalkyl-C₁-C₆alkoxy-C₁-C₆alkyl, C₁-C₆alkylcarbonyl-C₁-C₆alkyl,

C₁-C₆alkoxycarbonyl-C₁-C₆alkyl, C₁-C₆alkylthio, C₂-C₆alkenylthio, C₃-C₆alkynylthio, C₃-C₆cycloalkylthio, C₃-C₆cycloalkyl-C₁-C₆alkylthio, C₂-C₆haloalkenylthio, C₁-C₆haloalkylthio, NH₂, NH(C₁-C₆alkyl), N(C₁-C₆alkyl)₂, C₁-C₆alkylcarbonylamino, C₁-C₆haloalkylcarbonylamino, C₁-C₆alkoxycarbonylamino, C₁-C₆alkylaminocarbonylamino, -SO-C₁-C₆alkyl, -SO-halo-C₁-C₆alkyl, -SO₂-C₁-C₆alkyl, -SO₂-halo-C₁-C₆alkyl, -C(=O)R₁₁, phenyl or benzyl; wherein the phenyl and benzyl radicals may be unsubstituted or may carry independently of each other one to three substituents selected from the group consisting of halogen, OH, SH, CN, nitro, C₁-C₆alkyl, C₁-C₆haloalkyl, C₁-C₆alkylcarbonyl, C₂-C₆alkenyl, C₂-C₆haloalkenyl, C₂-C₆alkynyl, C₁-C₆alkoxy, C₁-C₆haloalkoxy, C₂-C₆alkenyloxy, C₂-C₆haloalkenyloxy, C₂-C₆alkynyoxy, -S(=O)-C₁-C₆alkyl, -S(O)₂-C₁-C₆alkyl, C₁-C₆alkoxycarbonyl and C₂-C₆haloalkenyloxy; and

R₁₁ is H, OH, C₁-C₆alkyl, C₃-C₈cycloalkyl, C₃-C₈cycloalkyl-C₁-C₆alkyl, C₁-C₆haloalkyl, C₁-C₆alkoxy, C₃-C₈cycloalkoxy, C₃-C₈cycloalkyl-C₁-C₆alkoxy, C₁-C₆haloalkoxy, C₂-C₆alkenyl, C₂-C₆haloalkenyl, C₂-C₆alkenyloxy, C₂-C₆haloalkenyloxy, C₂-C₆alkynyl, C₂-C₆haloalkynyl, C₂-C₆alkynyoxy, C₂-C₆haloalkynyoxy, NH₂, NH-C₁-C₆alkyl, -N(C₁-C₆alkyl)₂, NH-phenyl, NH-benzyl, phenoxy or benzyloxy;

and, where applicable, their possible E/Z isomers, E/Z isomeric mixtures and/or tautomers, in each case in free form or in salt form.

2. A compound according to claim 1 of formula (I) in free form.
3. A compound according to either claim 1 or claim 2 of formula (I), wherein X₁ and X₂ are chlorine or bromine.
4. A pesticidal composition which comprises as active ingredient at least one compound according to claim 1 of formula (I), in free form or in agrochemically acceptable salt form, and at least one adjuvant.
5. A process for the preparation of a composition as described in claim 4, which comprises intimately mixing the active ingredient with the adjuvant(s).
6. A method of controlling pests, which comprises applying a pesticidal composition as described in claim 4 to the pests or to the locus thereof.
7. Use of a compound according to any one of claims 1 to 3 of formula (I), in free form or, where applicable, in agrochemically acceptable salt form, in the preparation of a composition as described in claim 4.

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